University of Alberta

Impacts of reservoir creation on the biogeochemical cycling of methylmercury in boreal forest uplands

by

Britt Dianne Hall ©

A thesis submitted to the Faculty of Graduate Studies and Research in partial fulfillment of the

requirements for the degree of Doctor of Philosophy.

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Date submitted: August 13/03

'A lake is the landscape's most beautiful and expressive feature. It is earth's eye; looking into which the beholder measures the depth of his own nature. The fluviatile trees next the shore are the slender eyelashes which fringe it, and the wooded hills and cliffs around are its overhanging brows'.

'The forest has never so good a setting, nor is so distinctly beautiful, as when seen from the middle of a small lake amid hills which rise from the waters edge; for the water in which it is reflected not only makes the best foreground, but, with its winding shore, makes the most natural and agreeable boundary to it.'

-Henry David Thoreau
Walden, 1854

University of Alberta

Faculty of Graduate Studies and Research

The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies and Research for acceptance, a thesis entitled "Impacts of reservoir creation on the biogeochemical cycling of methylmercury in boreal forest uplands" submitted by Britt Dianne Hall in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Environmental Biology and Ecology.

Vincent St. Louis

David Schindler

Kevin Devito

Chris Le

Peter Dillon

Date approved: August 07/03

This work is dedicated to my grandparents,
Bill and Eva Rudd,
whose wisdom and love I will always treasure.

Abstract

An important consequence of reservoir creation is the production and bioaccumulation of neurotoxic methylmercury (MeHg) through the food web into fish. The FLooded Upland Dynamics EXperiment (FLUDEX) at the Experimental Lakes Area in NW Ontario was designed to test the hypothesis that MeHg production in reservoirs is related to the amount, and subsequent decomposition, of flooded organic matter. From 1999-2001, three upland forests that varied in the amounts of organic carbon stored (OC) in vegetation and soils (Low C, 30 900 kg C ha^{-1}; Medium C, 34 900 kg C ha^{-1}; and High C, 45 900 kg C ha^{-1}) were flooded from spring to autumn with low OC, low-MeHg water pumped from a near-by lake. Within the framework of the FLUDEX, this thesis examined whole-reservoir rates of methylation or demethylation based on net MeHg exports from reservoirs and MeHg pools in soils, periphyton, zooplankton, and fish. There was an initial pulse of MeHg production in all upland reservoirs in the first two years post flood (120 - 1590 ng m^{-2} day^{-1}), followed by net demethylation (360 - 1230 ng MeHg degraded m^{-2} day^{-1}) that reduced the pools of MeHg in the reservoirs. The reservoir with the highest amount of stored OC produced the most MeHg. Large increases in MeHg stores in soils (231 – 3230 mg ha^{-1}) compared to those in water (32 – 131 mg ha^{-1}) and biota (0.1 – 50 mg ha^{-1}) indicated that flooded soils were the main sites of methylation. Reservoirs were always net sources of MeHg to downstream environments (0.01 – 1.24 mg ha^{-1} day^{-1}).

This whole-ecosystem scale research was complemented with two smaller studies designed to examine the process of Hg methylation in reservoirs. Enclosure and three-year litterbag experiments simultaneously quantified production of MeHg and decomposition by-products following inundation of 12 plant tissues commonly found in boreal regions. MeHg production was greater in less easily decomposable tissues than in more labile tissues, contradictory to the hypothesis that the production of MeHg is primarily related to rates of OC decomposition. This study should assist hydroelectric utilities in making informed decisions about selecting sites for future reservoir development to reduce MeHg contamination of reservoir fisheries.

Preface

Collaboration with other scientists with expertise that complements one's own is one of the most rewarding aspects of whole ecosystem research. I am indebted to many individuals that have contributed ideas, data, editing, and enthusiasm over the course of my studies. In appreciation, I have given credit by way of authorship to those that were instrumental in the preparation of three manuscripts that have been, or will be, published from this thesis.

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Chapter 3: Hall, B.D. and V.L. St. Louis. Methylmercury and total mercury in plant litter decomposing in upland forests and flooded landscapes. *Environmental Science and Technology*. In preparation.

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☿

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Hintelmann, Marc Amyot, and Reed Harris, as well as many others I have interacted with over the course of my career, have all participated in generating exciting research, stimulating ideas, as well as help whenever I asked for it. A special thank you goes to John Rudd and Carol Kelly for always being interested in, enthusiastic about, and supportive of my work. As well, there were many people that contributed efforts in the field and lab and I have acknowledged them at the end of each chapter of this thesis.

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I am blessed with many wonderful friends, many of which I met during my tenure at the ELA. A heartfelt thanks to all of you who understand that life doesn't get any better than when you are paddling a canoe or sitting on a warm rock with people that make you laugh and think, and inspire all to insist on richness and meaning in our lives.

To my parents; Judy and Doug Mackay, Gerald Hall, and Alex and Susan Cudney, my sisters, Krista and Nancy, and their families, and rest of my talented, wonderful, dynamic family… thank you, thank you, thank you. And to finally to my beloved companions, Paul and Uisce, I am so grateful for our little family and am looking so forward to our next adventure!

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Chapter 1. General Introduction

Environmental impacts of reservoir creation

Large-scale flooding and water diversion projects have many negative environmental impacts (Rosenberg and others 2000; St. Louis and others 2000). Many of these projects that are constructed in the boreal forest produce hydroelectric energy for southern populations (ICOLD1998) and reduce biodiversity (Rosenberg and others 1995), lower ocean levels (Vorosmarty and Sahagian 2000), and potentially effect the rotation of the earth (Chao 1991, 1995). It has recently been shown that reservoirs are also sources of the greenhouse gases (GHGs) carbon dioxide (CO_2) and methane (CH_4) released to the atmosphere after production through microbial mineralization of flooded organic carbon (OC) in soils and vegetation (Rudd and others 1993; Duchemin and others 1995; St. Louis and others 2000; Matthews and others submitted).

An important consequence of reservoir creation is the bioaccumulation of methylmercury (MeHg), a strong vertebrate neurotoxin, through the food web into fish. Concentrations of MeHg in fish harvested from northern boreal reservoirs often exceed Canadian marketing limits of 0.5 $\mu g\ g^{-1}$ wet mass (Lodenius and others 1983; Bodaly and others 1984; Hecky and others 1991; Brouard and others 1994; Scruton and others 1994; Schetagne and Verdon 1999) and consumption of fish is the primary pathway of MeHg to humans (Hightower and Moore 2003). MeHg has a half-life of 44 days in humans (Smith and others 1994) and therefore the body burdens of MeHg in humans accumulates rapidly with relatively few dietary exposures per month (Mahaffey and Mergler 1998). Recently, the U.S. Environmental Protection Agency and National Academy of Science recommend maintaining blood mercury level of <5 $\mu g\ L^{-1}$, which corresponds to a daily intake of 0.1 μg kg body weight^{-1} day^{-1} (Hightower and others 2003). The health of people depending on reservoir fisheries for food becomes of concern (Rosenberg and others 1997), because human consumption of fish with sufficiently high concentrations of MeHg may cause teratogenic effects and irreversible neurological damage (Kurland and others 1960).

Biogeochemical cycle of mercury in natural forests

Mercury is a highly volatile metal that is easily transported from anthropogenic sources (mainly coal-fire electricity generation and waste combustion) to remote areas (Mason and Sheu 2002). The majority of mercury in the atmosphere is gaseous elemental Hg^0 which can be oxidized to the di-positive mercuric ion Hg^{+2} (HgII) in photo-catalytic reactions (Fitzgerald and others 1998). HgII is water-soluble and enters the terrestrial ecosystem either through direct precipitation or as dry deposition to the forest canopy, which is then deposited on the forest floor either as throughfall from the canopy during subsequent rain events or as litterfall (St. Louis and others 2001). HgII enters aquatic systems as direct deposition or contained in runoff from surrounding watersheds (Hurley and others 1995; Rudd 1995).

Once in anaerobic regions wetlands and lake sediments, HgII is biomethylated to MeHg primarily by sulphate reducing bacteria (Compeau and Bartha 1985). Biomethylation appears to be an enzyme-catalyzed process (Choi and others 1994) requiring the presence of three coenzymes, the most important being the cobalt-containing methyl-corrinoid B_{12} derivative methylcobalamin (Beijer and Jernelov 1979; Berman and others 1990). This biomethylation of inorganic Hg depends on the metabolism of the methylating organisms in anaerobic zones of reservoirs, as well as the amount of substrates, such as HgII, small organic compounds, and sulphate ($SO_4^{=}$) bioavailable for methylation. As well, the extent of Hg methylation may be affected by changes in environmental factors that have been shown to directly affect methylation, such as pH, anoxia (Winfrey and Rudd 1990; Gilmour and Henry 1991), temperature (Bodaly and others 1993), and selenium concentrations (Rudd and Turner 1983; Turner and Rudd 1983; Fjeld and Rognerud 1993). The production of MeHg is reversible either by microbial demethylation (Robinson and Tuovinen 1984) or ultra-violet photo-degradation (Sellers and others 1996). Demethylation reduces HgII to Hg^0 which can then be dissipated to the atmosphere (Winfrey and others 1990). The atmosphere is also a source of MeHg to terrestrial and aquatic systems (Fitzgerald and others 1998; Rolfhus and others 2003). In some remote forest, atmospheric deposition generally contributes relatively insignificant amounts of MeHg (St. Louis and others 2001). However, boreal plants contain MeHg, possibly from uptake from soils and soil waters or from methylation inside plant tissues. During leaf drop, this MeHg in plants is an important input of MeHg to forest floors (St. Louis and others 2001).

Generally, lakes and upland forests tend to be strong sinks for total Hg (THg, an analytical term used to describe all forms of Hg) and MeHg, both of which can accumulate in the organic and mineral soils and lake sediments (St. Louis and others 1996; Sellers and others 2001). Unperturbed wetlands are generally sources of MeHg and small sinks for THg (St. Louis and others 1996). When upland forest and wetland areas are inundated in the creation of reservoirs, the decomposition of flooded OC in anoxic zones in reservoirs also fuels the microbial methylation of HgII to MeHg (Compeau and Bartha 1984; Hecky and others 1991; Kelly and others 1997) and the resulting landscape becomes a source of both MeHg and inorganic Hg to downstream environments.

Reservoir studies at the Environmental Lakes Area

The amount of MeHg produced in reservoirs may be dependent upon the amount and type of flooded OC available for mineralization. For example, the quantity of easily decomposed labile carbon flooded may be very important to the short-term decomposition processes fuelling MeHg and GHG production. To begin examining the link between decomposition, GHG production, and Hg methylation in reservoirs, a unique multidisciplinary, whole-ecosystem experiment was initiated at the Experimental Lakes Area (ELA; Fig. 1-1) in 1990/91 (Kelly and others 1997). The ELA Reservoir Project (ELARP) flooded a wetland complex, composed of a 14.4 ha peatland surrounding a 2.4 ha pond, after two years of monitoring in its natural state. A wetland was chosen for the ELARP because it was thought to provide the worst-case scenario for long-term MeHg and GHG production due to the large stores of OC in peat available for decomposition over the long-term.

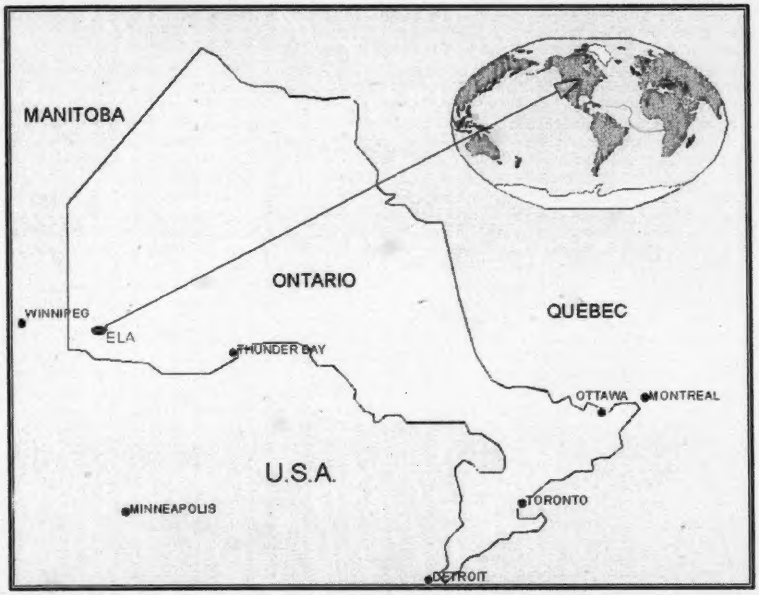

Figure 1-1. The location of the Experimental Lakes Area. The ELA is a set of boreal lakes designated by the Canadian Federal and Ontario Provincial governments for limnological and ecological research. The ELA research station is located 50 km southeast of Kenora, Ontario, on the Precambrian Shield. (Brunskill and Schindler 1971).

Within a few weeks of flooding, the production of MeHg and GHG increased dramatically (Kelly and others 1997). Concentrations of MeHg after flooding were approximately 10 times greater in water (Kelly and others 1997), 2-10 times greater in invertebrates (Hall and others 1998), and 2-3 times greater in fish (Bodaly and others 1997) than prior to flooding. Continued background monitoring of dissolved MeHg, CO_2, and CH_4 in the wetland reservoir showed that rates of MeHg and GHG production were well above background levels nine years after initial flooding (St. Louis and others submitted).

Results from the ELARP led to the hypothesis that minimizing the percentage of peatland flooded should, in the long term, reduce the production of MeHg and subsequent bioaccumulation into fish. This hypothesis was addressed by initiating a new whole ecosystem flooding experiment at the ELA in 1998. The FLooded Upland Dynamics Experiment (FLUDEX) flooded three upland boreal forest sites that varied in amounts of OC stored in soils and vegetation. The three upland reservoirs (called Low C, Medium C, and High C; Figs. 1-2, 1-3, and 1-4) contained between 30 900 – 45 900 kg C ha^{-1}, substantially less OC than that stored in the wetland reservoir (1.26 million kg C ha-1). The upland reservoirs were flooded annually in May/June from 1999-2001 with low OC carbon, low-MeHg water pumped from a nearby lake (Fig. 1-5). A major objective of the FLUDEX was to determine if flooding upland and wetland landscapes that vary in the amount of OC flooded results in similar increases in MeHg production. This whole ecosystem manipulation experiment was complimented with smaller studies designed to examine some of the Hg biogeochemical processes occurring in upland reservoirs.

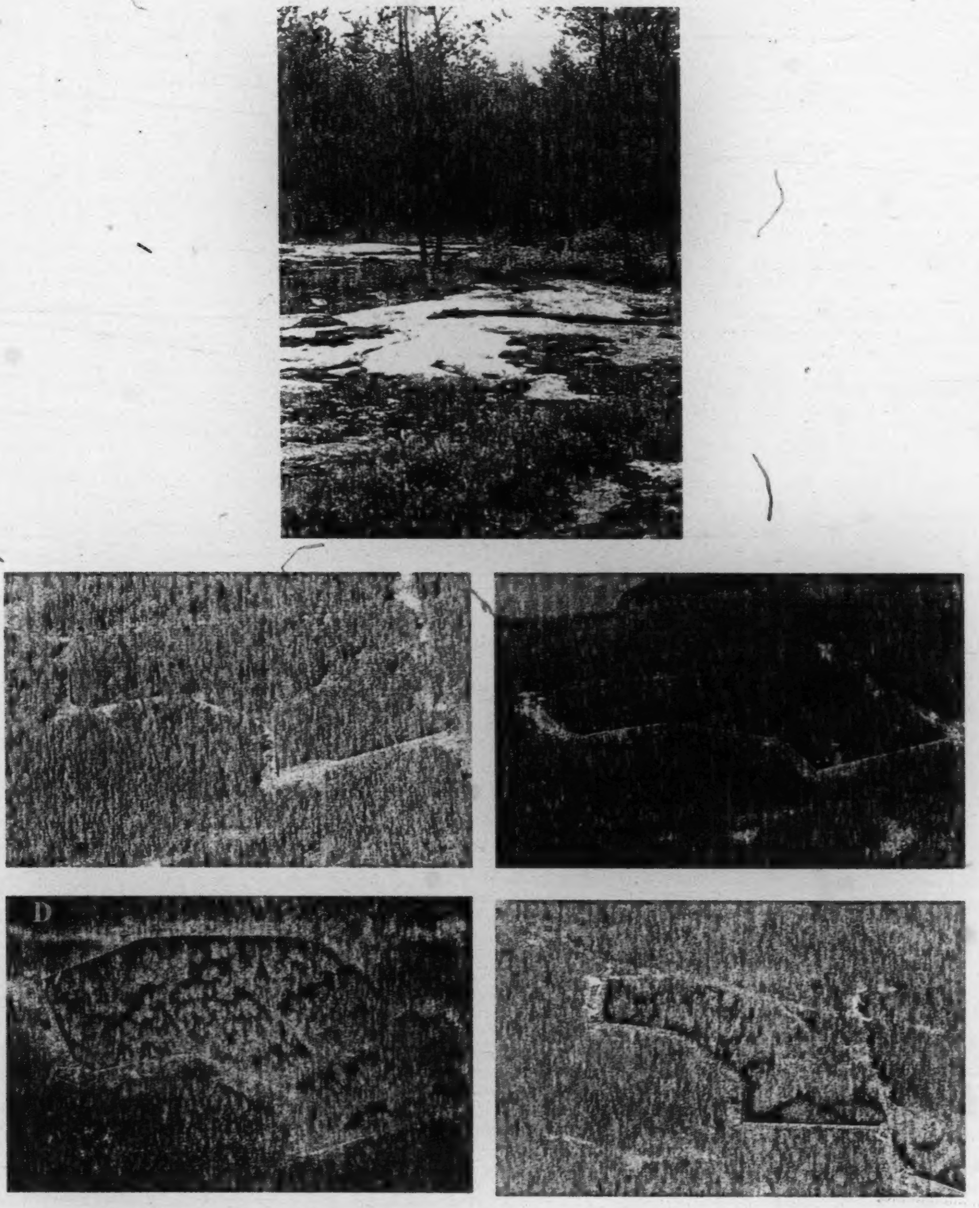

Figure 1-2. The Low C reservoir. A. Ground view of the site prior to flooding. B-E. Aerial view of the reservoir before flooding in 1998 (B), and one (C), two (D), and three (E) years post flooding.

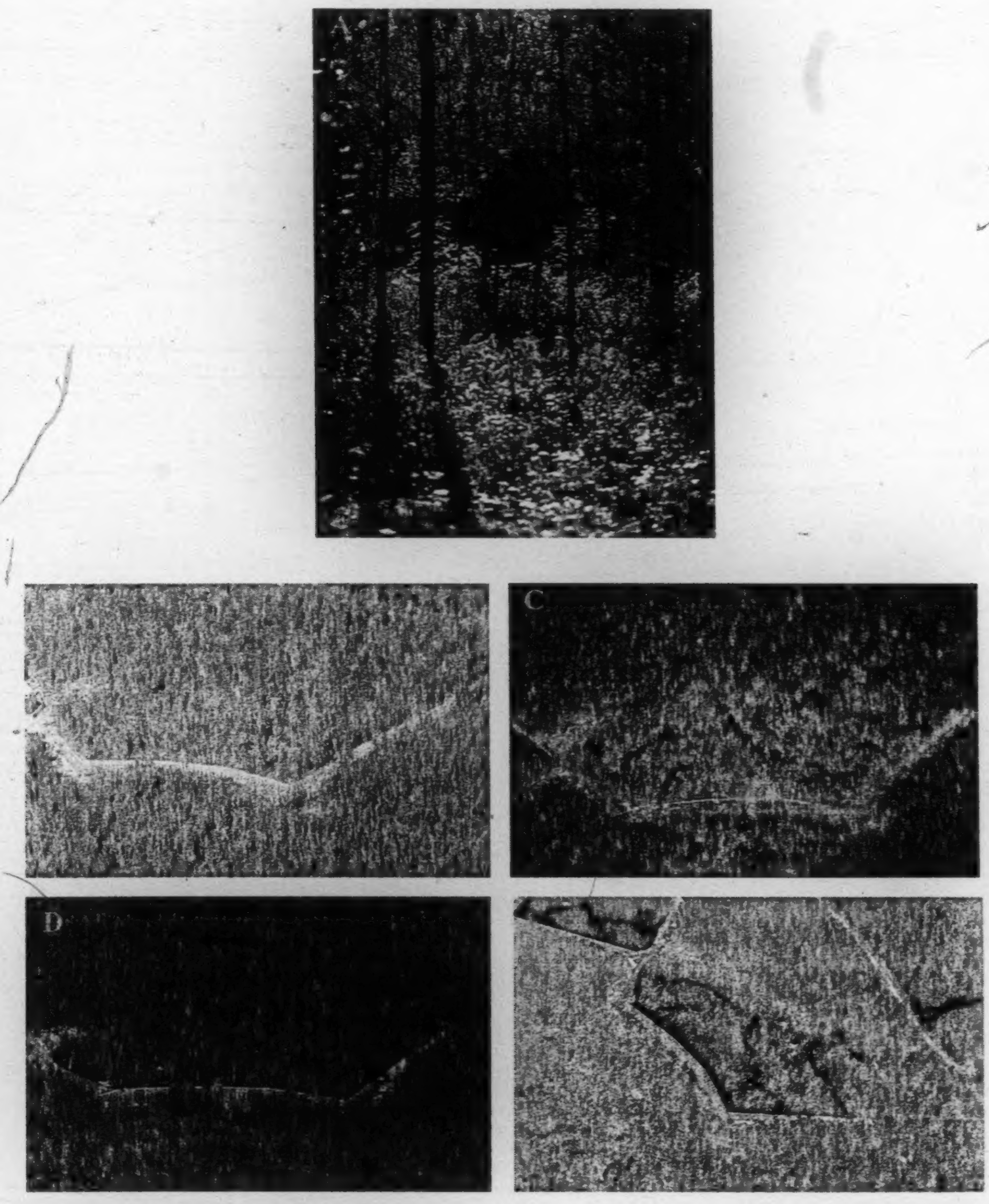

Figure 1-3. The Medium C reservoir. A. Ground view of the site prior to flooding. B-E. Aerial view of the reservoir before flooding in 1998 (B), and one (C), two (D), and three (E) years post flooding.

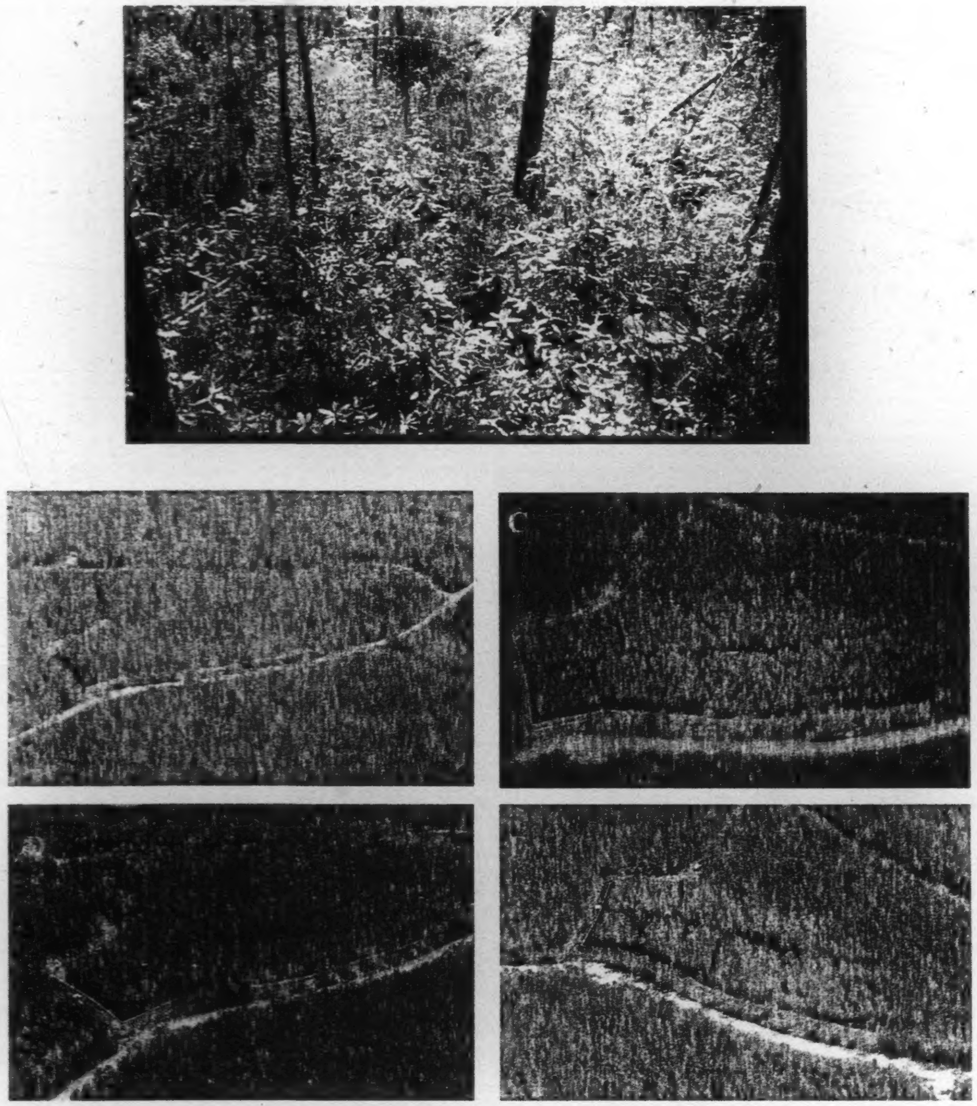

Figure 1-4. The High C reservoir. A. Ground view of the site prior to flooding. B-E. Aerial view of the reservoir before flooding in 1998 (B), and one (C), two (D), and three (E) years post flooding.

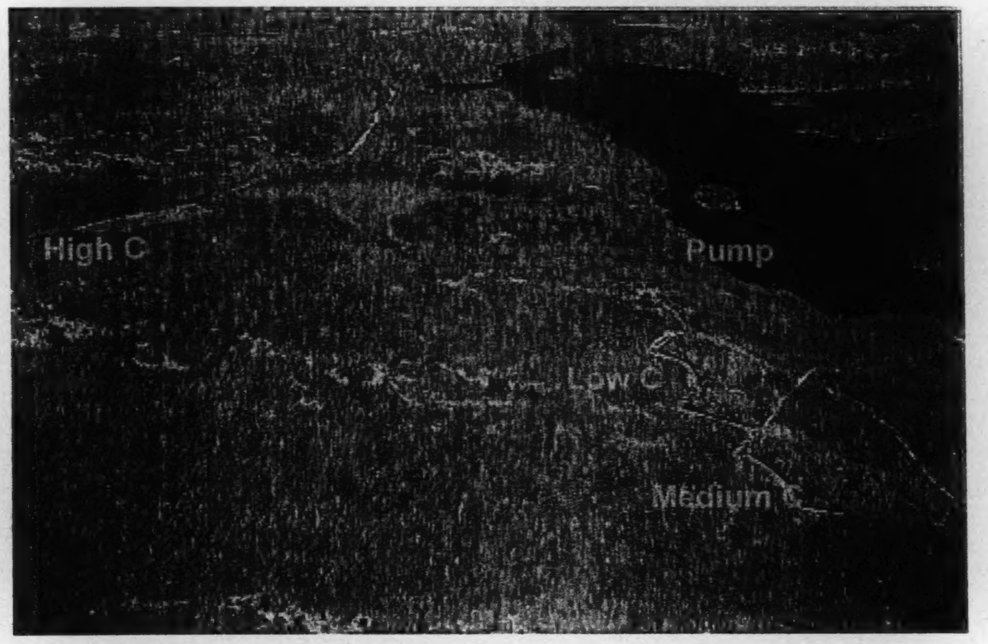

Figure 1-5. Aerial view of all three upland reservoirs and location of the inflow pump. (Photo credit: C.J.D. Matthews).

Within the framework of the FLUDEX, the overall goals of this thesis are to quantify the production of MeHg in upland reservoirs and to determine whether the net production of MeHg in reservoirs is related to the amount of OC flooded. An auxiliary goal is to examine processes that contribute to increased methylation rates in upland reservoirs. This thesis consists of three research chapters. Chapter 2 describes an enclosure experiment that simultaneously quantified the production of MeHg and decomposition by-products following inundation of two plant tissues (birch [*Betula papyrifera*] leaves and jack pine [*Pinus banksiana*] needles) commonly found in boreal regions where northern reservoirs are created. Birch leaves decomposed 2.4 times faster than jack pine needles as measured by the total carbon decomposition by-products produced in enclosures over time. However, measured net MeHg production in enclosures containing birch leaves (0.35±0.05 ng per g carbon added) was five times lower than in the enclosures containing jack pine needles (1.94±0.28 ng per g carbon added). These results showed that MeHg production is not solely related to rates of OC decomposition, and that increases in MeHg associated with flooded birch leaves and jack pine needles resulted from the production of new MeHg as opposed to leaching of MeHg already in the plant tissues during decomposition.

Chapter 3 describes a three-year litterbag study examining rates of MeHg production, THg leaching, and decomposition of 12 boreal plant tissues placed in the High C, Medium C, and Low C reservoirs and in unflooded forested sites adjacent to each of the upland reservoirs. The results of this study showed that, while THg decreased in some decomposing plants due to leaching, substantial increases in THg mass were observed in other tissues. Plant tissues placed in reservoirs exhibited large increases in MeHg mass, whereas plant tissues placed in unflooded forests lost MeHg mass over time.

Chapter 4 is the primary focus of this thesis and examines the impact of reservoir creation on the biogeochemical cycling of MeHg and THg in boreal upland forests. MeHg production in each upland reservoir was determined by estimating the total MeHg storage in water, soils, and biota. Results from the upland reservoirs are compared to the flooded wetland complex. There was an initial pulse of MeHg production in all reservoirs that lasted for 2 years, after which net demethylation began to reduce the pools of MeHg in the reservoirs. Rates of methylation and demethylation were generally related to the amount of total OC stored in the reservoirs before flooding.

Finally, a concluding chapter presents a general summary of the thesis, as well as a discussion of possible avenues of future research.

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Chapter 2. The stimulation of methylmercury production by decomposition of flooded birch leaves and jack pine needles

Introduction

Concentrations of the neurotoxin methylmercury (MeHg; CH_3Hg^+) in fish in reservoirs often exceed Health Canada consumption guidelines of 0.5 $\mu g\ g^{-1}$ wet mass (Brouard and others 1994; Scruton and others 1994; Bodaly and Fudge 1999). Reservoirs are also sources of the greenhouse gases carbon dioxide (CO_2) and methane (CH_4) to the atmosphere due to aerobic and anaerobic microbial mineralization of organic carbon in flooded soils and vegetation (Rudd and others 1993; Duchemin and others 1995; St. Louis and others 2000; Matthews and others submitted). Decomposition of flooded organic matter in reservoirs has been hypothesized to fuel the production of MeHg, which bioaccumulates in aquatic food webs, resulting in fish with elevated Hg concentrations.

A number of studies utilizing field (Roulet and others 2000; Roulet and others 2001; Balogh and others 2002; Porvari and others 2003), litterbag (Heyes and others 1998, Chapter 3, this thesis), mesocosm (Hecky and others 1991), and laboratory (Porvari and Verta 1995; Thérien and Morrison 1999) approaches have shown that inundation of plant tissue results in increased MeHg concentrations in surrounding water and biota. Whole ecosystem flooding experiments at the Experimental Lakes Area (ELA) in northwestern Ontario, Canada, examining changes in the biogeochemical cycling of carbon and Hg in response to reservoir creation (Kelly and others 1997; Chapter 4, this thesis; St. Louis and others submitted), have also demonstrated that decomposition of flooded organic matter in reservoirs resulted in increased MeHg concentrations in food web organisms. These findings have spurred debate focusing on two possible mechanisms that may cause these elevated MeHg levels.

The first mechanism is the simple leaching during decomposition of MeHg already present in flooded organic matter. As a result, the rate at which MeHg enters the food web may be directly proportional to the rate at which organic matter is mineralized. Although THg and MeHg concentrations in boreal plants are typically very low (Moore and others 1995; Munthe and others 1995; Rasmussen 1995), litterfall has been shown to be an important input of MeHg and THg to forest floors (St. Louis and others 2001). MeHg in organic matter has been associated with low molecular weight water-soluble fulvic acids (Reddy and Aiken 2001), and therefore may be more leachable than inorganic forms of Hg (Hultberg and Munthe 2001). Under this scenario, the quantity of MeHg that could enter the reservoir food web would never exceed what was stored in organic matter prior to flooding.

The second mechanism involves the production of new MeHg following reservoir creation (Kelly and others 1997). This biomethylation of inorganic Hg depends on the metabolism of the methylating organisms (e.g., sulphate reducing bacteria; SRB) in anaerobic zones of reservoirs, as well as the amount of substrates, such as HgII, small organic compounds, and sulphate ($SO_4^{=}$) bioavailable for methylation metabolism. Rates of Hg methylation may therefore be directly proportional to rates of microbial mineralization of organic carbon because products of decomposition are important substrates for Hg methylation. The extent of Hg methylation may be affected following flooding by changes in environmental factors that have been shown to directly affect methylation, such as pH (Winfrey and Rudd 1990; Gilmour and Henry 1991), temperature (Bodaly and others 1993), selenium (Se) concentrations (Rudd and Turner 1983; Turner and Rudd 1983; Fjeld and Rognerud 1993), and anoxia. In this scenario, the amount of MeHg available for bioaccumulation can far exceed what was initially there in organic matter, and the production of new MeHg can persist for long periods of time following flooding.

Although past studies have shown that flooding organic carbon results in increases in MeHg in water and biota, these studies were unable to determine if increases in MeHg were due to leaching or new MeHg production, nor did they concurrently measure increases in MeHg and decomposition by-products of flooded plant tissue. Here we describe an enclosure experiment in which we simultaneously quantify the production of MeHg and decomposition by-products following inundation of two plant tissues (birch (*Betula papyrifera*) leaves and jack pine (*Pinus banksiana*) needles) found in the boreal ecoregion where northern reservoirs are commonly created. We hypothesized that: 1. there will be production of new MeHg associated with the decomposition of flooded plant tissues, and 2. rates of MeHg production will be directly proportional to rates of decomposition.

Methods

Experimental set up

Six rigid, opaque plastic enclosures were placed randomly in a raft floating in a sheltered area of an oligotrophic lake at the ELA (Lake 240) for 53 days in July 2000. Each enclosure was filled with ~420 L of water from Lake 240, which contained low concentrations of both dissolved organic carbon (146 μmol L^{-1}) and MeHg (0.02 ng L^{-1}), leaving a headspace of ~75 L (Fig. 2-1). Two enclosures each received 300 g of either fresh birch leaves or fresh jack pine needles hand picked while wearing vinyl gloves. Two enclosures contained only lake water. Please see Fig. 2-1 for further details of enclosure setup.

Sampling protocol and analytical methods

By-products of decomposition

- Carbon in plants

To quantify the amount of carbon added as plant tissue to each treatment enclosure, carbon content of freeze-dried birch leaves and jack pine needles was analyzed at the beginning and end of the experiment using an Exeter Analytical Model 440 Elemental Analyzer at the University of Alberta Limnology Laboratory. The relationship between fresh and dry weights was determined by weighing sub-samples of plant tissue before and after freeze-drying. The mass of carbon added to each enclosure was determined by multiplying the dry weight of plant tissue by the % carbon content.

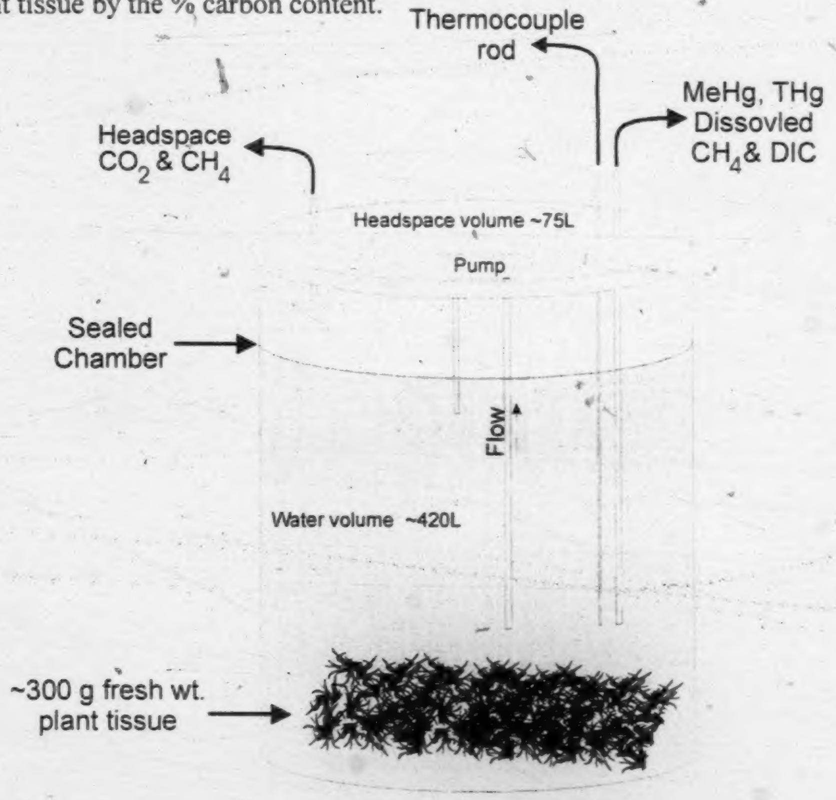
Thermocouple rod
Headspace CO_2 & CH_4
MeHg, THg Dissovled CH_4& DIC
Headspace volume ~75L
Pump
Sealed Chamber
Flow
Water volume ~420L
~300 g fresh wt. plant tissue

Figure 2-1. A cross-section of an enclosure containing plant tissue. Each enclosure was ~75 cm wide and 115 cm deep, and filled with ~420 L of whole lake water leaving a ~75 L headspace. Lids for each enclosure contained a Teflon tube with an acid cleaned nitex mesh filter for water sampling and a Constantan copper-nickel thermocouple rod attached to a Campbell Scientific datalogger for monitoring water temperatures. Lids were attached to enclosures using silicone caulking. The water in each enclosure was mixed for 20 minutes prior to sampling by pumping bottom water into the headspace using high volume water pumps equipped with acid-rinsed Teflon gears and fittings (St. Louis and others 2003).

- Inorganic carbon in water and enclosure headspace

We measured increases in concentrations of dissolved inorganic carbon (DIC) and dissolved CH_4 (by-products of organic carbon mineralization) in enclosures over time. The opaque enclosures prevented any photosynthetic assimilation of CO_2 produced during mineralization of plant tissue. Samples for dissolved gas analyses were collected twice a week. Just prior to sampling, the water in each enclosure was mixed for 20 minutes by pumping bottom water into the headspace using high volume water pumps.

Dissolved gas samples were collected in evacuated Wheaton bottles of known volume (~160 mL) containing 8.9 g of KCl and backfilled with 10 mL of ultra high purity (UHP) nitrogen (N_2) to maintain a headspace. 0.5 mLs of concentrated phosphoric acid was injected into each sample to convert all bicarbonate to CO_2. A volume of Lake 240 water was injected back into enclosures to replace that removed in samples. Enclosure headspace samples were collected into evacuated 60 mL Wheaton bottles through a septum in the lid using double-ended needles.

Samples containing dissolved gases were shaken on a wrist action shaker for 10 minutes prior to analysis to equilibrate dissolved CO_2 and CH_4 with the N_2 headspace. A 0.2 mL sample of headspace from the water samples, as well as the enclosure headspace, was injected into a Varian Model 3800 gas chromatograph (GC) using a Hamilton pressure lock syringe. Gas samples were analyzed using a flame ionization detector at 250°C and UHP hydrogen as a carrier gas. Chromatographic separation was achieved through a Hayes Sep D column at 80°C. A ruthenium methanizer converted CO_2 to CH_4. Standards (Praxair; Linde-Union Carbide) ranging from 20 to 19900 CO_2 ppm and 1.6 to 78.1 CH_4 ppm, were used to generate calibration curves ($r^2>0.98$) before and after each set of samples. Peak areas were integrated using Star WorkStation analytical software. CO_2 and CH_4 were converted to concentrations (µmol L^{-1}) using Henry's Law (Hamilton and others 1994), correcting for temperature and barometric pressure differences between sample collection and gas analysis.

- Water chemistry including organic carbon

To characterize differences in the chemical composition of the water in the enclosures, water samples for general chemistry were taken in Nalgene polypropylene bottles from each enclosure at the beginning and end of the experiment. Samples were analyzed at the ELA Aquatic Analytical Chemistry Laboratory for parameters that might affect potential Hg methylation in aquatic systems, such as pH, alkalinity, and dissolved ions (Cl^-, $SO_4^=$, K^+, Mg^{+2}, Ca^{+2}, and Fe^{+2}) (Stainton and others 1977). Other products of decomposition, such as dissolved and suspended nutrients, were also measured. Suspended carbon and dissolved organic carbon (DOC) concentrations were divided by the amount of carbon added as plant tissues to standardized differences in suspended carbon and DOC among treatments.

- Total carbon pools

We estimated rates of decomposition of plant tissues by calculating the total amount of carbon produced in each enclosure. The total carbon pool dissolved in the water in each enclosure was calculated by multiplying DIC, CH_4-C, DOC, and suspended C concentrations by the water volume in each set of enclosures. The total inorganic carbon pool in the headspace was calculated by multiplying CO_2 and CH_4-C concentrations by the headspace volume. The average total carbon pool in enclosures containing only water was subtracted from the pool in enclosures containing birch leaves or jack pine needles and divided by the amount of carbon in plant tissues added to determine standardized carbon production among treatments. Inorganic carbon pools (DIC, CO_2, and CH_4) were calculated for each sampling date, whereas total carbon including DOC and suspended C were calculated at the end of the experiment only.

Mercury

- Mercury in plants

Both MeHg and THg concentrations were measured in plant tissues to: 1) assess the amount of MeHg and THg added to each enclosure at the beginning of the experiment, and 2) determine if there was a net increase in MeHg in the plant tissues over the course of the experiment. A sub-sample of plant tissues added to the enclosures was freeze-dried and ground using an acid-rinsed stainless steel coffee grinder. At the end of the experiment, after removing the majority of the water from enclosures, a portion of the decomposed plant tissue was collected by pouring the remaining water through a 500 μm mesh. Plant samples were analyzed for concentrations of MeHg and THg at Flett Research Ltd. (Winnipeg, Manitoba). 5-6 mg of freeze-dried plant tissues was digested in 300 μL KOH/MeOH overnight at 75°C and analyzed for MeHg using cold vapour atomic fluorescence spectrophotometry (CVAFS) after ethylation (Bloom 1989; Horvat and others 1993; Liang and others 1994). For THg analysis, plant tissues were first digested in 1 HNO_3:2.5 H_2SO_4 for 6 hours at 250°C. Digests were cooled, diluted with ultra-clean distilled water, and BrCl was added to maintain all Hg species in solution as HgII. Following digestion, samples were analyzed using CVAFS (Bloom and Crecelius 1983; Bloom and Fitzgerald 1988). Detection limits were 0.25 ng g^{-1} for MeHg and 1 ng g^{-1} for THg.

The mass of MeHg and THg added to enclosures in fresh plant tissues was calculated by multiplying the dry weight of tissues by the concentration of MeHg and THg. The decomposed plant tissue in the enclosures (especially birch leaves) became suspended in the water column over the course of the experiment, and only a small amount could be collected at the end of the experiment to determine concentrations of MeHg and THg. Unfortunately, final plant weight could not be determined and a final mass of MeHg in the decomposing plant tissues could not be measured directly. We therefore used a THg mass balance budget to estimate final plant weights assuming that, because the enclosures were closed systems, the initial and final mass of THg in the water and plants in each enclosure did not change over the course of the experiment even though some THg may have leached from plant tissues during decomposition. We solved for the final THg mass in plant tissues as follows (known quantities are italicized):

(*Initial THg mass in water*) + (*Initial THg mass in plant tissues*) = (*Final THg mass in water*) + (Final THg mass in plant tissues);

where the THg mass in water was calculated by multiplying the volume of the enclosures by the concentration of THg in the water at the beginning and end of the experiment, and the initial THg mass in plant tissues was calculated by multiplying the concentration of THg in birch leaves and jack pine needles by the amount of plant tissue added on a dry weight basis to each enclosure. We then solved for the final weight of plant tissues:

(Final weight of plant tissues) = (*Final THg mass in plant tissues*) / (*Final THg concentration in plant tissues*)

The final plant weights were multiplied by final MeHg concentrations in plants to determine the MeHg mass in jack pine needles and birch leaves at the end of the experiment.

- Mercury in water

Unfiltered enclosure water was collected for MeHg and THg analyses through Teflon lines attached to a high volume water pump fitted with acid rinsed Teflon gears. Samples were pumped into ultra-clean 125 or 250 ml Teflon bottles using clean-hands-dirty-hands sampling protocol (St. Louis and others 1996). Flett Research Ltd. analyzed unfiltered water samples for MeHg and THg concentrations. Twenty percent of samples were taken in duplicate. MeHg samples were frozen until analysis and THg samples were preserved using trace metal grade HCl (to 1% of total sample volume). Samples were analyzed for MeHg using CVAFS after distillation (Horvat and others 1993) and aqueous phase ethylation (Bloom 1989) (method detection limits = 0.015 ng L^{-1} at a blank level of 0.02-0.03 ng L^{-1}). Total Hg samples taken in the first two weeks and at the end of the experiment were analyzed by using CVAFS as described in (Bloom and others 1988) (method detection limit of 0.1 ng L^{-1} at a blank level of 0.3-0.4 ng L^{-1}). The total MeHg mass in the water in each enclosure at the beginning and end of the experiment was calculated by multiplying MeHg concentrations by the water volume in enclosures.

- Net increases in methylmercury mass

The final total mass of MeHg in each enclosure was calculated by adding the mass of MeHg in the plant tissues to the mass of MeHg in the water. The net increase in MeHg mass in treatment enclosures was calculated as follows:

(Final MeHg mass in water and plant tissues) - (Initial MeHg mass in water and plant tissues) - (Average increase in MeHg mass in water in control enclosures).

A standardized rate of MeHg production per mass of carbon added in plant tissue was determined by dividing the final net increase in MeHg mass by the mass of carbon in plant tissues added.

Results

By-products of decomposition

- Carbon in plants

The percent dry weight and carbon content of fresh jack pine needles (49.1±3.7% and 52.0±0.1% carbon, respectively; Table 2-1) were greater than those in fresh birch leaves (34.2±3.0% and 49.3±0.3% carbon, respectively). As a result, there was 1.5 times more carbon added to the enclosures containing jack pine needles (76.6±5.9 g carbon) than to the enclosures containing birch leaves (50.7±4.9 g carbon; Table 2-1).

Table 2-1. Weight and % carbon content in plant tissues. Values for individual enclosures are presented in parentheses.

	Fresh plant tissue added (g)	Equivalent dried mass added[1] (g)	%C in dried plants at the start of the experiment	C added (g)	% C in dried plants at end of experiment
Birch leaves	300±0 (300, 300)	103±8.9 (93.8, 112)	49.3±0.5 (48.9, 49.8)	50.7±4.9 (45.8, 55.5)	55.4±0.1 (55.3, 55.4)
Jack pine needles	300.0±0 (300, 300)	147±11.1 (136, 158)	52.0±0.1 (51.9, 52.1)	76.6±5.9 (70.7, 82.5)	53.8±0.1 (53.7, 53.8)

[1] calculated using ratio of fresh weight to dry weight ratios

- Inorganic carbon in water and enclosure headspace

We initially sealed the headspace of the enclosures from the atmosphere with silicon caulking under the lids. However, this seal was broken sometime between days 15 and 33. On Day 33, when the leak was detected, enclosures were resealed with additional silicon caulking. Despite the leaks, however, it was evident that birch leaves decomposed faster than jack pine needles. The average concentrations of DIC in treatment enclosures began to increase immediately after plant tissues were added, and were always higher than average DIC concentrations in the enclosures containing only water (Fig. 2-2A). Average DIC concentrations in enclosures containing birch leaves exceeded those in enclosures with jack pine needles after Day 33 (ANOVA; p=0.002; F=82.75), reaching a maximum of 1190 μmol L^{-1} at the end of the experiment. Average DIC concentrations in enclosures containing jack pine needles increased at the beginning of the experiment, but plateaued at concentrations between 500 and 600 μmol L^{-1} from Day 19 on. Average DIC concentrations in enclosures

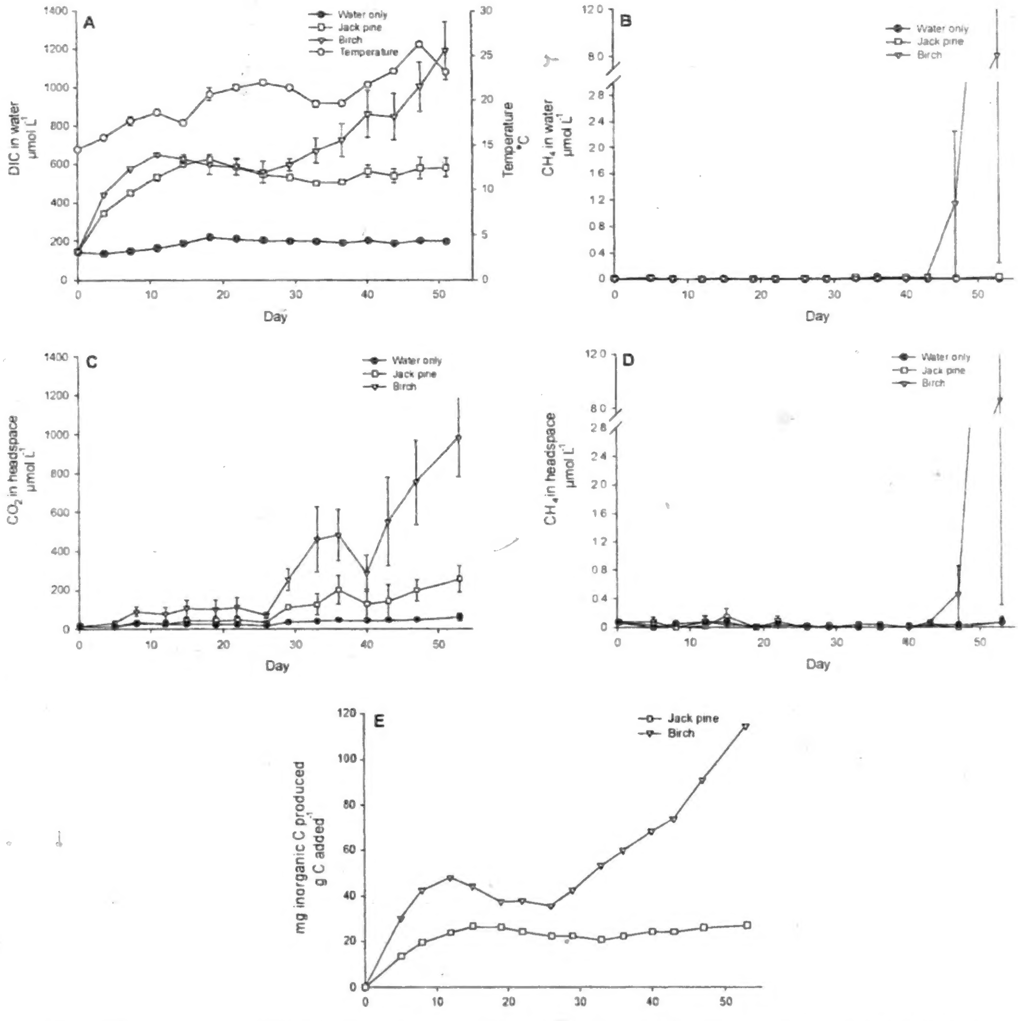

Figure 2-2. Concentrations of A: dissolved inorganic carbon and B: dissolved methane in unfiltered water and C: carbon dioxide and D: methane in the headspace from enclosures containing jack pine needles, birch leaves, and water only. E: Total mass of inorganic carbon produced in unfiltered water per gram of carbon added as either jack pine needles or birch leaves. Total inorganic carbon produced was calculated by subtracting the C produced in the control enclosures from the total C in unfiltered water and headspace of each enclosure and dividing by the mass of C in dried plants at the beginning of the experiment (Table 2-1). Inorganic C production per g C added was calculated for each enclosure and then averaged.

CO_2 concentrations were significantly higher in enclosures containing birch leaves compared to those containing jack pine needles (ANOVA; p=0.006, F= 43.326). Increases in CO_2 concentrations in the headspace of the jack pine barrels ranged from 15 to over 250 μmol L^{-1} and were less pronounced than increases in enclosures containing birch leaves, which reached concentrations of 980 μmol L^{-1} by the end of the experiment (Fig. 2-2B). Average headspace CO_2 concentrations in the control barrel never exceeded 65 μmol L^{-1}. There was virtually no CH_4 detected in either the enclosure water or headspace, with the exception of one containing birch leaves, which had elevated CH_4 concentrations in both the water and the headspace after Day 43 of the experiment (Fig. 2-2C and D).

- Water chemistry including organic carbon

Water chemistry data also suggested that birch leaves decomposed faster than jack pine needles. Concentrations of total suspended and dissolved nutrients and some cations (Ca^{+2}, K^{+}, Mg^{+}) at the end of the experiment were higher in enclosures containing birch leaves than in enclosures containing jack pine needles (Table 2-2). Concentrations of SO_4 in the water of enclosures with jack pine needles were lower than enclosures with birch leaves at the end of the experiment (Table 2-2), possibly suggesting greater rates of SO_4 reduction there than in enclosures with birch leaves. pH was lower in the treatment enclosures compared to the control enclosures (Table 2-2) and alkalinity was higher in enclosures containing jack pine needles than in enclosures with birch leaves. There were no changes in dissolved concentrations of Na^{+} (0.90-0.94 mg L^{-1}), Fe^{+2} (0.01-0.07 mg L^{-1}), and Cl^{-} (0.28-0.47 mg L^{-1}) among enclosures over the duration of the experiment (Table 2-2). There were no differences in DOC concentrations in enclosures with birch leaves compared to those with jack pine needles, however, the average suspended carbon concentration was 1.9 times

Table 2-2. Concentrations of chemical constituents in water from enclosures at the beginning and end of the enclosure experiment. Averages represent data from two replicate enclosures ± one standard error.

Chemical Parameter	Initial	Final		
		Birch	Jack pine	Control
Nutrients[1]				
DOC (μmol L^{-1})	585±15	1650±240	1620±50	595±5
TDN (μg L^{-1})	310±45	1045±425	425±15	310±5
NH_4^+ (μg L^{-1})	32±19	1215±1195	20±2	59±24
NO_2^- (μg L^{-1})	0±1	0	0	0
$NO_3^=$ (μg L^{-1})	1±2	1±0	0	3±2
TDP (μg L^{-1})	3.5±0.5	7.0±3.0	5.5±3.5	8.0±0
Suspended C (μg L^{-1})	610±50	6895±455	3580±1320	435±125
Suspended N (μg L^{-1})	49±4	1188±114	478±153	47±15
Suspended P (μg L^{-1})	4±0	240±21	73±26	3±1
Chlorophyll *a* (μg L^{-1})	1.90±0.66	0.19±0.08	0.14±0.05	0.08±0.02
Physical parameters				
pH	7.23±0.02	5.70±0.06	5.84±0	6.61±0.13
Alkalinity (μeq L^{-1})	138.5±1.5	374.5±6.5	214.5±7.5	140.0±3.0
Conductivity (μS cm^{-1})	25.5±0.5	45.0±3.0	29.5±0.5	26.5±0.5
Anions				
Cl^- (mg L^{-1})	0.28±0.6	0.35±0.015	0.47±0.1	0.35±0.03
$SO_4^=$ (mg L^{-1})	2.74±0.33	0.94±0.01	0.38±0.09	2.79±0.04
Cations				
Fe^{+2} (mg L^{-1})	0.05±0.01	0.07±0.03	0.01±0.06	0.01±0.02
Ca^{+2} (mg L^{-1})	2.34±0.09	3.49±0.03	2.26±0.02	2.41±0.01
Na^+ (mg L^{-1})	0.92±0.01	0.94±0	0.92±0.01	0.90±0.01
Mg^+ (mg L^{-1})	0.76±0.02	1.44±0.05	0.98±0.01	0.76±0.01
K^+ (mg L^{-1})	0.43±0.01	2.36±0.04	1.59±0.04	0.43±0.01

[1] DOC= dissolved organic carbon
TDN= total dissolved N
TDP= total dissolved P

higher in enclosures with birch leaves compare to those in enclosures with jack pine needles (Table 2-2).

- Total carbon pools

The amount of inorganic carbon produced (*i.e.*, mineralized) per g carbon in plant tissue was 1.4 to 3.9 times higher in enclosures with birch leaves than in enclosures with jack pine needles (Fig. 2-2E). Flooded birch leaves produced a maximum of 97.5 mg inorganic carbon per g carbon added by the final day of the experiment (Fig. 2-2E). Flooded jack pine needles produced a maximum of 26.5 mg inorganic carbon per g carbon added by Day 12. After Day 12, it appears that mineralization ceased because the amount of inorganic carbon produced per g carbon added remained relatively constant between 19.6 and 26.0 mg for the remainder of the experiment. Because enclosures were not properly sealed for a period of approximately 18 days in the middle of the experiment, a portion of the CO_2 and CH_4 produced was lost to the atmosphere. If the amount of leakage was dependent on the rates of inorganic C production, we would have observed greater loss of gaseous C from the enclosure containing birch leaves, which supports other observations of increased decomposition in flooded birch leaves compare to jack pine needles.

At the end of the experiment, the average DOC pools were similar among the treatment enclosures, however average suspended carbon pools were 1.9 times greater in the enclosures with birch leaves (Fig. 2-3A). The total carbon pool, including all inorganic and organic forms of carbon, was 1.3 times greater in the enclosures with birch leaves compared to those containing jack pine needles. Once carbon pools were standardized to the amount of carbon added as plant tissue, the enclosures with birch leaves produced 269 mg per g C added as plants, which was 2.3 times greater than the total carbon produced per g C added as plants in the enclosures with jack pine needles (110 mg per g C added; Fig. 2.3B).

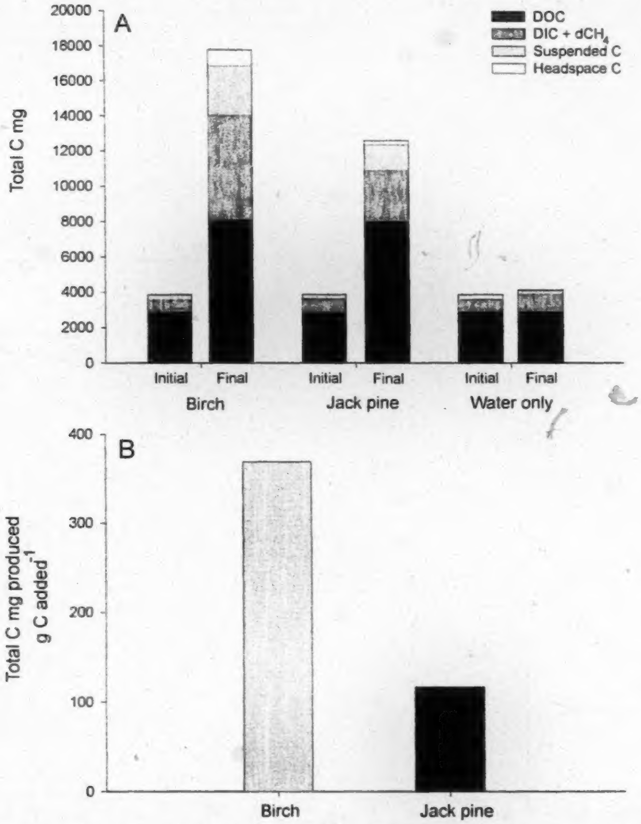

Figure 2-3. A: Initial and final average total carbon pools in enclosures including dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), dissolved methane (dCH_4), suspended carbon, and carbon in enclosure headspace. B: Total carbon produced at the end of the 53-day experiment, standardized to the amount of carbon added in plant tissue.

Mercury

- Mercury in plants

MeHg concentrations in fresh birch leaves (0.36 ng g^{-1}) were 3.6 times higher than in jack pine needles (0.10 ng g^{-1}). THg concentrations in fresh birch leaves (9.31 ng g^{-1}) were 1.3 times lower than in jack pine needles (12.26 ng g^{-1}; Table 2-3). At the end of the experiment, average MeHg and THg concentrations in birch leaves (0.69±0.28 ng MeHg g^{-1}, 45.29 ± 9.86 ng THg g^{-1}) were ~2 fold higher than jack pine needles (0.32±0.15 ng MeHg g^{-1}, 19.01±3.47 ng THg g^{-1}; Table 2-3).

The percent THg that was MeHg (%MeHg) in fresh plant tissues was initially much higher in birch leaves (3.87%) than in jack pine needles (0.82%). %MeHg in birch leaves decreased over the course of the experiment to 1.45±0.29, while %MeHg in jack pine needles increased to 1.70±0.23 (Table 2-3). At the end of the experiment, there was no difference in %MeHg between tissue types (Table 2-3).

Table 2-3. Concentrations of methylmercury (MeHg) and total mercury (THg), and percent THg that was MeHg in plant tissues. Values for individual enclosures are presented in parentheses.

	Birch leaves			Jack pine needles		
	THg (ng g^{-1} d.w.)	MeHg (ng g^{-1} d.w.)	%MeHg	THg (ng g^{-1} d.w.)	MeHg (ng g^{-1} d.w.)	%MeHg
Initial	9.31	0.36	3.87	12.26	0.10	0.82
Final	45.28±9.86 (55.13, 35.42)	0.69±0.28 (0.96, 0.41)	1.45±0.29 (1.74, 1.16)	19.01±3.47 (22.49, 15.54)	0.32±0.15 (0.33, 0.30)	1.70±0.23 (1.47, 1.93)

The jack pine needles (final modelled mass = 82.5 and 118.6 g d.w.) lost 31.7 ± 12.2% of their original mass, whereas the birch leaves (final modelled mass = 18.8 and 34.2 g d.w.) lost 74.2±7.5% of their mass by the end of the 53-day experiment. But the calculation of plant mass at the end of the experiment was only an estimate. However, decomposition as indicated by the dissolved inorganic carbon concentrations was 1.4 to 3.9 times faster in flooded birch leaves than in flooded jack pine needles. This is similar to differences in final mass of plant tissues modelled using THg concentrations, which show that birch leaves decomposed approximately 2.4 times faster than jack pine needles.

The modelled final mass of MeHg in jack pine needles (31.4±4.2 ng MeHg) was ~2 times greater than the modelled final MeHg mass of birch leaves (16.1±2.0 ng MeHg) (Table 2-4).

Table 2-4. Methylmercury (MeHg) mass in unfiltered water and plant tissue at the beginning and end of the experiment. Values for individual enclosures are presented in parentheses.

	Initial MeHg (ng)			Final MeHg (ng)			Net MeHg increase[2]
	Plants	Water	Total	Plants[1]	Water	Total	
Birch	36.8±0.12 (37.0, 36.7)	8.24±0.04 (8.24, 8.24)	45.1±0.08 (45.1, 45.1)	16.1±2.03 (18.1, 14.0)	81.2±4.50 (76.7, 85.7)	97.3±2.48 (94.8, 99.8)	17.6±2.57 (15.0, 20.1)
Jack pine	14.9±0.14 (14.7, 15.0)	8.20±0 (8.16, 8.24)	23.1±0.14 (23.0, 23.2)	31.4±4.18 (27.2, 35.6)	175±25.8 (201, 149)	206±21.6 (228, 185)	149±21.7 (170, 127)
Control	0	8.20±0.12 (8.32, 8.08)	8.20±0.12 (8.32, 8.08)	0	42.8±0.39 (42.5, 43.2)	42.8±0.39 (42.5, 43.2)	

[1] Final plant MeHg mass values were calculated from modelled final plant weights (see text).

[2] (Final MeHg mass in water and plants) – (initial MeHg mass in water and plants) - (average increase in MeHg mass in water in control enclosures).

- Mercury in water

Average concentrations of MeHg in water were significantly higher in enclosures with jack pine needles (0.42±0.06 ng L-1) than in enclosures with birch leaves (0.20±0.01 ng L-1) at the end of the experiment (ANOVA $p = 0.004$, $F = 57.90$; Tukey's test $p = 0.021$) (Fig. 2-4A). Average concentrations of MeHg in water in enclosures with plant tissues were significantly higher than concentrations in enclosures with only water by Day 29 (ANOVA; $p = 0.002$, $F = 88.51$) and remained so until the end of the experiment.

Average THg concentrations in water decreased slightly at the beginning of the experiment; however, by the end of the experiment, THg concentrations were not significantly different from initial concentrations (ANOVA; $p= 0.515$, $F = 0.84$) (Fig. 2-4B). In all enclosures with plant tissues, the %MeHg in water was greater at the end of the experiment compared to the beginning (Fig. 2-4C). The %MeHg in water flooding jack pine needles (23.4±3.6) was 1.7 times higher than in water flooding birch leaves (13.5±2.5) at the end of the experiment, and 3.3 times higher than in enclosures containing only water (7.1±1.0).

At the end of the experiment the mass of MeHg in the water in enclosures with jack pine needles (175±25.8 ng) was ~2 times greater than in the water of enclosures with birch leaves (81.2±4.50 ng) (Table 2-4). There was a 5-fold increase in MeHg mass (42.8± 0.39 ng) in enclosures with only water over the duration of the experiment.

- Net increases in methylmercury mass

The mass of MeHg in water was similar among enclosures at the beginning of the experiment (~8 ng; Table 2-4). However, there was more MeHg added to enclosures in birch leaves (36.8±0.1 ng) than in jack pine needles (14.9±0.1 ng) (Table 2-4). As a result, the initial total MeHg mass in the enclosures with birch leaves (45.1±0.1 ng) was ~2 times greater than in the enclosures with jack pine

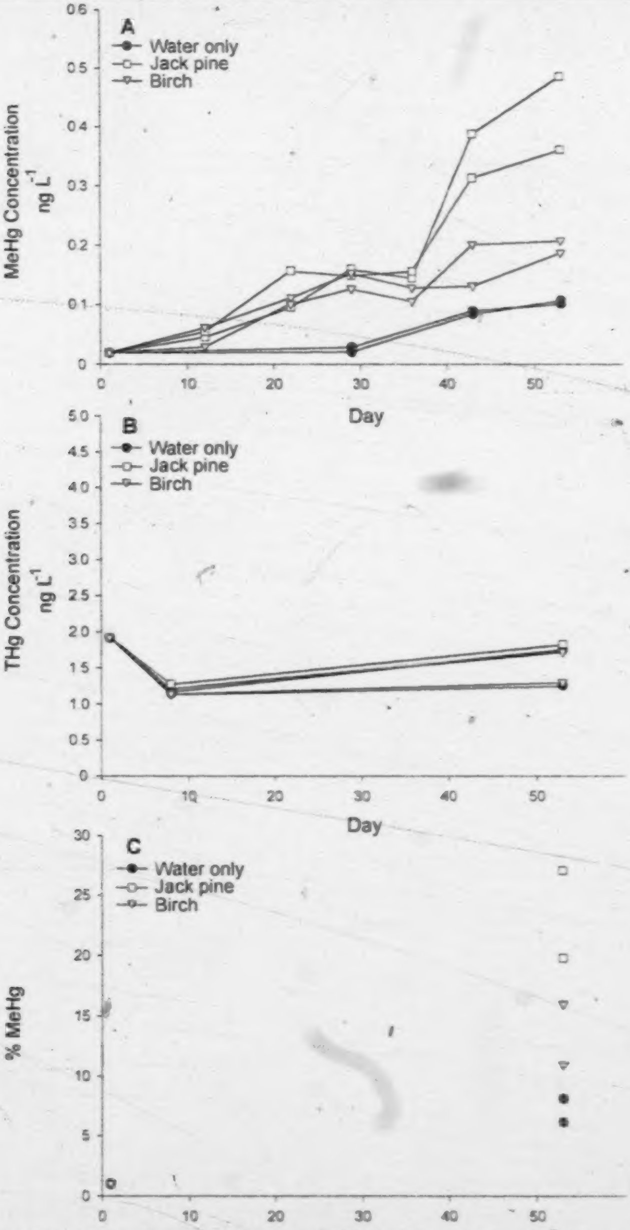

Figure 2-4. A: Methylmercury concentrations (ng MeHg L^{-1})in unfiltered water from enclosures containing jack pine needles, birch leaves, and water. B: Total mercury concentrations (ng THg L^{-1}) in unfiltered water from enclosures containing jack pine needles, birch leaves, and water. C: The percent of THg that was MeHg in unfiltered water.

needles (23.1±0.1 ng) (Fig. 2-5). At the end of the experiment, total mass of MeHg in water and plant tissues was 206.4±21.6 ng and 97.3±2.5 ng for enclosures with jack pine needles and birch leaves, respectively (Fig. 2-5).

The average *total net increase* of MeHg mass in the jack pine needles treatment (149±21.7 ng) exceeded that in the birch leaves treatment (17.6±2.57 ng) by over 8 fold, despite greater initial MeHg mass in the birch treatment (Table 2-4) and higher rates of decomposition. The average ng MeHg produced per g carbon added in plant tissues, as calculated by dividing the net MeHg increase in each enclosure by the amount of C added in dried plants, was almost 5 times higher in enclosures containing jack pine needles (1.94±0.28) compared to enclosures containing birch leaves (0.35±0.05) (Fig. 2-6).

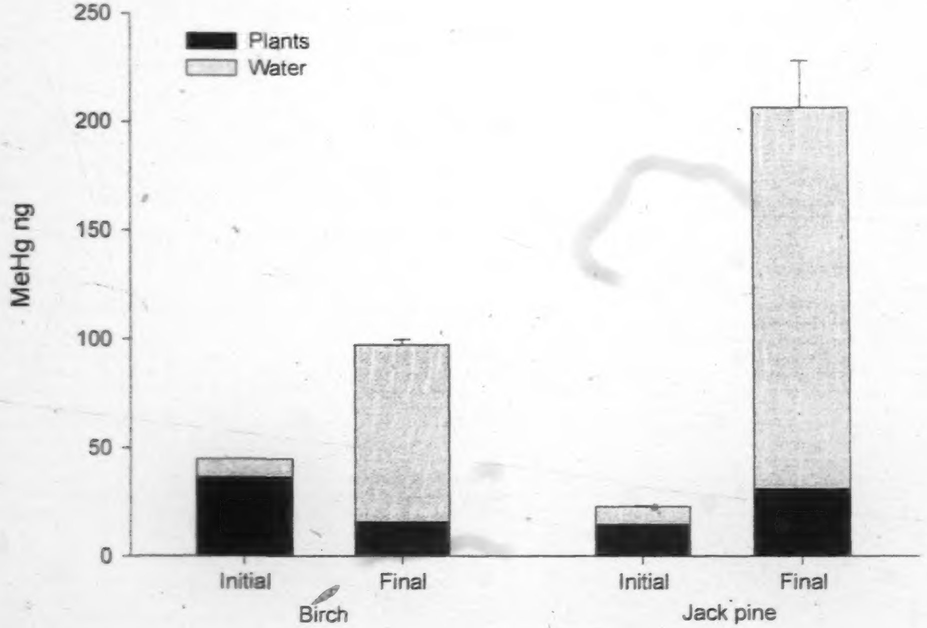

Figure 2-5. Total initial and final methylmercury mass (ng MeHg) in unfiltered water and plants in treatment enclosures.

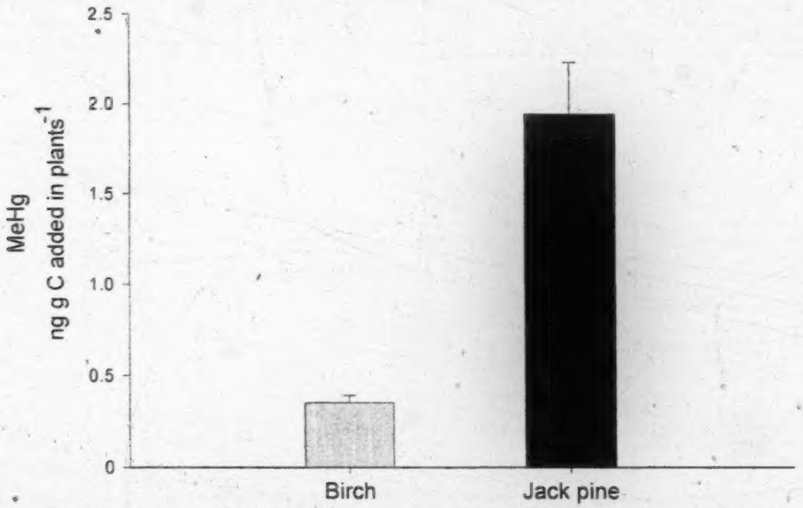

Figure 2-6. Net mass of methylmercury (MeHg) produced per mass of carbon added as either jack pine needles or birch leaves. Net MeHg production was calculated dividing net MeHg increases (Table 2-3) by the mass of C in dried plants at the beginning of the experiment (Table 2-1). MeHg production per g C added was calculated for each enclosure and then averaged.

Discussion

This study addressed two hypotheses: 1. there will be production of new MeHg associated with the decomposition of flooded plant tissues, and 2. rates of MeHg production will be directly proportional to rates of decomposition. The increases in MeHg mass clearly show that net methylation was stimulated by flooding jack pine needles and birch leaves. However, there was 8 times less MeHg production associated with flooded birch leaves compared to flooded jack pine needles, despite birch leaves decomposing ~2.4 times faster than jack pine needles. While our data support our first hypothesis that there was production of new MeHg associated with the decomposition of flooded plant tissues (as opposed to just leaching of MeHg already present in tissues), the data did not support our second hypothesis that rates of Hg methylation are directly proportional to rates of organic carbon mineralization.

Decomposition

Decomposition of flooded birch leaves was ~2.4 times greater than flooded jack pine needles as indicated by modelled mass loss of tissues, concentrations of DIC, DOC, and suspended carbon, as well as concentrations of other products of decomposition such as suspended and dissolved nutrients. The large increases in DIC and low pH in the water column suggest that the majority of inorganic C in the water column existed as dissolved CO_2 (Hutchinson 1957), however, there was also a likely increase in the amount of organic acids released from the plant tissues during decomposition. Increases in alkalinity in water containing birch leaves and jack pine needles were likely due to the release of positively charged species (NH_4^+ and base cations) from plant tissue during decomposition, coupled with the decrease in anions due to increased rates of SO_4 reduction (Dillon and others 1997). Studies in streams in Spain (López and others 2001) and in the Swedish boreal ecoregion (Haapala and others 2001) also found that birch (*Betula* sp.) leaves decomposed more rapidly than other plant tissues such as Monterey pine (*Pinus radiata*), *Eucalyptus nitens*, ray grass (*Lolium perenne*), and willow (*Salix* sp.) leaves when placed in water.

Unfortunately, we did not measure dissolved oxygen or redox potential in our enclosures, however, concentrations of dissolved oxygen would most likely have been negatively correlated with decomposition and therefore lower in the enclosures with birch leaves. The general absence of CH_4 in the water and headspace of our enclosures also indicates that large zones of anoxia did not form and that decomposition in the water column was aerobic. O_2 was required for aerobic decomposition, which indicates that there was a substantial leak of O_2 into the enclosures. However, we cannot estimate the O_2 in the water column because the enclosures were re-sealed and we cannot be certain that the enclosure headspace was at equilibrium with the atmosphere.

Methylmercury production vs. leaching

The amount of MeHg added to the enclosures as birch leaves was almost double that added to enclosures as jack pine needles, yet at the end of the experiment, the mass of MeHg in water flooding jack pine needles was 3.4 times higher than MeHg mass in water flooding birch leaves. When the net measured production of MeHg was standardized to the amount of carbon added in plants, the amount of MeHg produced per g carbon added in the jack pine treatments (1.94±0.28 ng) exceeded that in birch treatments (0.35±0.05 ng) by 5 times over the course of the experiment. An increase in the %MeHg often indicates increased net methylation rates within aquatic systems (Kelly and others 1995; Rudd 1995). The %MeHg in all of our enclosures increased by the end of the experiment, but the average %MeHg in water containing jack pine needles was almost double that in the water containing birch leaves. If leaching had been the primary source of MeHg to water in our enclosures, the mass of MeHg in the water in the treatment enclosures would not have exceeded the initial mass of MeHg in the enclosures.

Why did we observe more methylation associated with the decomposition of jack pine needles than with the decomposition of birch leaves?

Our experimental results support the hypothesis that there was production of new MeHg as a result of the decomposition of flooded plant tissues as opposed to leaching from decomposing flooded organic matter. The results of our study are consistent with other studies that examined MeHg increases in coniferous needles compared to deciduous leaves and grasses. Studies in the South Indian Lake reservoir in northern Manitoba demonstrated that when spruce (*Picea mariana*) boughs were added to enclosures containing perch, THg concentrations in these perch were greater than in those held in enclosures to which prairie sod and moss-peat were added (Hecky and others 1991). Black spruce needles sampled from litterbags placed in an experimentally flooded wetland at the ELA exhibited an increase of 800% of original MeHg mass, compared to increases of 630% of original mass in *Sphagnum fuscum* moss and 50% of original mass in sedge grass (*Carex rostrata*) stalks (Heyes and others 1998). Our results do not support our hypothesis of higher production of MeHg in the more easily decomposable birch leaves relative to jack pine needles.

We propose three possible explanations as to why there was greater production of MeHg associated with flooded jack pine needles compared to flooded birch leaves: 1. the bioavailability of HgII (i.e., the ability of HgII to pass through microbial cell walls into methylating organisms) differed among treatments; 2. environmental factors and biogeochemical processes affecting methylation may have differed among enclosures, and/or 3. rates of demethylation differed among treatments.

1. Bioavailability of HgII

Differences in MeHg production associated with decomposing plant tissues may be attributable to differing ability of HgII to enter methylating organisms. This *bioavailability* could be affected by differences in concentrations of DOC, which can bind to HgII resulting in Hg complexes that are, because of either ionic nature or size, unable to cross cell membranes (Benoit and others 2001; Haitzer and others 2002). Barkay et al. (1997) measured the bioavailability of HgII using a genetically altered bacterium that produced light when HgII crossed the cell membrane (Selifonova and others 1993). They found a negative relationship between the amount of DOC present and HgII bioavailability. In our study, at the end of the experiment there was 1.5 times more DOC in enclosures with birch leaves than in enclosures with jack pine needles. This possibly resulted in more inhibition of methylation in the birch leaves compared to jack pine needle treatments. The type and quality of DOC (not measured in our study) may also have been an important factor.

2. Environment factors affecting methylation

In our study, higher net MeHg production in the enclosures containing jack pine needles could indicate that environmental factors favoured methylation there over enclosures with flooded birch leaves. Stimulation of MeHg production has been found in environments with low pH (Winfrey and others 1990; Gilmour and others 1991), increased temperature (Bodaly and others 1993), decreased Se concentrations (Rudd and others 1983; Turner and others 1983; Fjeld and others 1993), low redox potential (*i.e.,* anoxia) (Compeau and Bartha 1984; Björnberg and others 1988), and increased SO_4 concentrations (Compeau and Bartha 1985; Gilmour and others 1992; Branfireun and others 1999).

The pH in water in both treatment enclosures was lower than in enclosures containing only water (Table 2-2), indicative of decomposition of flooded plant tissue (Schlesinger 1997). However, differences in average pH between treatments with birch leaves (5.7±0.06) and jack pine needles (5.85±0) were negligible. There were also no differences in water temperatures among enclosures (Fig. 2-2). This indicated that neither pH nor water temperature were important factors in differing methylation rates. Concentrations of Se were not measured in our enclosures; however, other studies have shown that Se concentrations are generally extremely low at the ELA (V. Palace and R.A. Bodaly, unpublished data), and therefore unlikely to suppress methylation.

Laboratory studies have shown that methylating SRB perform best in low redox environments (Compeau and others 1984; Björnberg and others 1988; Regnell and Tunlid 1991; Pak and Bartha 1998), and rates of methylation measured in vessels containing flooded organic matter were higher under anaerobic conditions than aerobic conditions (Porvari and others 1995). Despite the aerobic water column, anoxia could have occurred in micro-zones surrounding plant tissues while the enclosures sat undisturbed between sampling periods. Jack pine needles did not break into small pieces and float in the water column as the birch leaves did, and therefore anaerobic micro-zones were more likely to occur among jack pine needles sitting on the bottom of the enclosures, than in pieces of birch leaves floating in the aerobic water column.

Methylation of HgII has been shown to be stimulated by inputs of SO_4 to boreal peatlands (Branfireun and others 1999), and freshwater (Gilmour and others 1992) and estuarine (Compeau and others 1985) sediments, as might be expected if SRB are the environmentally relevant methylating bacteria. At the end of our experiment, concentrations of SO_4 in the water of enclosures with jack pine needles were lower than enclosures with birch leaves. Alkalinity in water containing jack pine needles was much higher than in water containing birch leaves. Both of these findings suggest more active SO_4 reduction, and hence HgII methylation, in enclosures with jack pine needles than in enclosures with birch leaves, supporting our hypothesis that anaerobic micro-zones may have formed around the jack pine needles.

3. Demethylation

Rates of methylation and demethylation cannot be measured directly without adding tracer mercury species (Ramlal and others 1986; Hintlemann and others 2000), and as a result, we are only able to determine *net* production of MeHg using a mass budget approach in the enclosures. Differences in the measured net amount of MeHg produced among treatments could be attributed to differences in demethylation rates. For example, if the organic carbon added to the birch enclosures stimulated rates of HgII methylation, but also enhanced rates of demethylation, we would expect to see less *net* methylation in the flooded birch leaves compared to flooded jack pine needles. It is possible that rates of both methylation and demethylation were increased in the enclosures containing birch

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leaves, resulting in lower *net* methylation in the enclosures containing birch leaves compared to those with jack pine needles where possibly only HgII methylation was stimulated.

Summary

The addition of fresh jack pine needles and birch leaves to our enclosures stimulated the production of new MeHg. The results of our experiment corroborate laboratory, microcosm, and field studies examining increases in MeHg concentrations water inundating plant tissue (Hecky and others 1991; Porvari and others 1995; Thérien and others 1999; Balogh and others 2002) and support our hypothesis that increased MeHg observed in flooded environments is due to the production of new MeHg and not the leaching of MeHg already present in flooded organic matter. There was no direct link between rates of decomposition and rates of measured net MeHg production. Differences in MeHg production associated with flooded birch leaves and jack pine needles could be due to differences in environmental factors affecting HgII bioavailability (e.g., binding to DOC), SO_4 reduction rates, and/or rates of demethylation. Our study suggests that the amount of organic carbon stored in a reservoir prior to flooding is not a good predictor of the extent of future MeHg increases. Reservoirs created by flooding upland forest that contains relatively less organic carbon stores may result in contamination of reservoir fisheries equal to, or exceeding, reservoirs created over wetland areas with very large organic carbon stores. Our study also suggests that litterfall inputs of jack pine needles into lakes and streams could stimulate Hg methylation more than litterfall inputs of birch leaves during the annual fall senescence.

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Chapter 3. Methylmercury and total mercury in plant litter decomposing in upland forests and flooded landscapes

Introduction

Mercury (Hg) is a highly volatile metal that is easily transported from anthropogenic sources to remote areas (Mason and Sheu 2002). Contamination of aquatic ecosystems with Hg creates health concerns because consumption of fish is the primary means by which humans are exposed to the neurotoxic, methylated form of Hg (methylmercury; MeHg; Hightower and Moore 2003). Therefore, a solid understanding of how Hg moves from the atmosphere through watersheds is crucial to understanding the contamination of fisheries.

Oxidized, inorganic Hg (HgII) can enter watersheds in a number of ways. HgII is water-soluble and is transferred from the atmosphere to watersheds directly in wet precipitation (St. Louis and others 1995). Reactive gaseous and/or particulate HgII can also be deposited to the forest canopy as dry deposition and washed off during precipitation events in throughfall. In the boreal ecoregion of Canada, litterfall inputs of Hg(II) to watersheds were actually three times higher than Hg(II) inputs in wet deposition (St. Louis and others 2001). Although it is not fully understood how the Hg(II) in foliage gets there, it is currently assumed that a large portion of it originates through stomatal uptake of atmospheric Hg^0 (Gustin and others 2000). During subsequent senescence and litter decomposition, HgII may be released (Heyes and others 2000), which can in turn be transferred through the watershed in runoff to aquatic systems (Hurley and others 1995; Rudd 1995; Babiarz and others 1998). Once in the anaerobic zones of wetlands (St. Louis and others 1994; Branfireun and others 1999), lake sediments (Gilmour and others 1992), or even saturated upland soils (Hultberg and Munthe 2001; Porvari and others 2003), HgII can be methylated to MeHg, primarily by sulphate reducing bacteria (Compeau and Bartha 1985), which then bioaccumulates through aquatic food webs (Bodaly and others 1997).

Although wet deposition can be an important source of HgII to watersheds far from anthropogenic sources, atmospheric deposition of MeHg to our study site (the Experimental Lakes Area (ELA) in northwestern Ontario) is relatively insignificant (St. Louis and others 2001; Chapter 4, this thesis). However, boreal plants contain MeHg, which is possibly obtained through uptake from soils and soil waters (Bishop and others 1998) or by methylation of Hg(II) inside plant tissues (Moore and others 1995). Litterfall can therefore be an important MeHg input to forest floors following senescence (Bishop and others 1998; St. Louis and others 2001). In well-drained upland soils, MeHg may be destroyed by microbial demethylation and evaded back to the atmosphere as reduced, elemental Hg^0. However, saturated soils promote production of MeHg (Heyes and others 2000), which can be mobilized in runoff and transported to nearby aquatic environments.

Generally, upland forest soils act as sinks for atmospheric inputs of total Hg (THg; all forms of Hg) and MeHg (St. Louis and others 1994) because the majority of THg and MeHg binds to organic and mineral soil particles. However, when terrestrial and wetland areas are inundated in the creation of reservoirs, the decomposition of flooded organic carbon in anoxic zones fuels Hg(II) methylation, and the resulting landscape becomes a source of both MeHg and Hg(II) to downstream environments (Hecky and others 1991; Kelly and others 1997; Chapter 4, this thesis; Balogh and others 2002). Previous studies at the ELA have shown that increased rates of bioaccumulation of MeHg in fish in reservoirs results from increased production of MeHg stimulated by the decomposition of organic matter in reservoirs, not from the leaching of MeHg already stored in the decomposing organic matter (Chapter 2, this thesis; Heyes and others 2000).

One of the objectives of this study was to examine the leaching of THg from, and the production of MeHg in, decomposing tissues from plants common to boreal upland forests at the ELA. We used litterbags to study THg, MeHg, C, and N cycling in 12 different plant tissues placed in three unperturbed boreal forest sites over an ~800 day period. Because litterfall represents such an important flux of Hg to watersheds, understanding THg leaching and MeHg production in decomposing litter in natural forests will help identify sources of MeHg contamination of fish in lakes.

An equally important focus of this study is to explore the contribution of different decomposing plant tissues to THg leaching and MeHg production in reservoirs. This study is part of the FLooded Upland Dynamics EXperiment (FLUDEX), a whole ecosystem manipulation designed to examine the biogeochemical cycling of Hg and carbon in three upland boreal forest reservoirs created over forests that varied in amounts of organic carbon stored in soils and vegetation (Chapter 4, this thesis; Matthews and others 2003; Venkiteswaran and others submitted). Reservoirs (with maximum depth of 2 m and average depth of 1 m) were constructed by building dikes along low-lying contours of the sites followed by flooding with water pumped from a nearby oligotrophic lake. The three reservoirs (called Low C, 30 900 kg C ha^{-1}; Medium C, 34 900 kg C ha^{-1}; and High C, 45 900; Fig. 3-1) were flooded annually in May or June from 1999-2001 with low-carbon, low-Hg water (THg and MeHg concentrations ~1.0 and 0.03 ng L^{-1}, respectively; see Chapter 4, this thesis). The reservoirs were emptied each September or October to simulate drawdown in the shallow zones of northern hydroelectric reservoirs in the winter due to increased power demand.

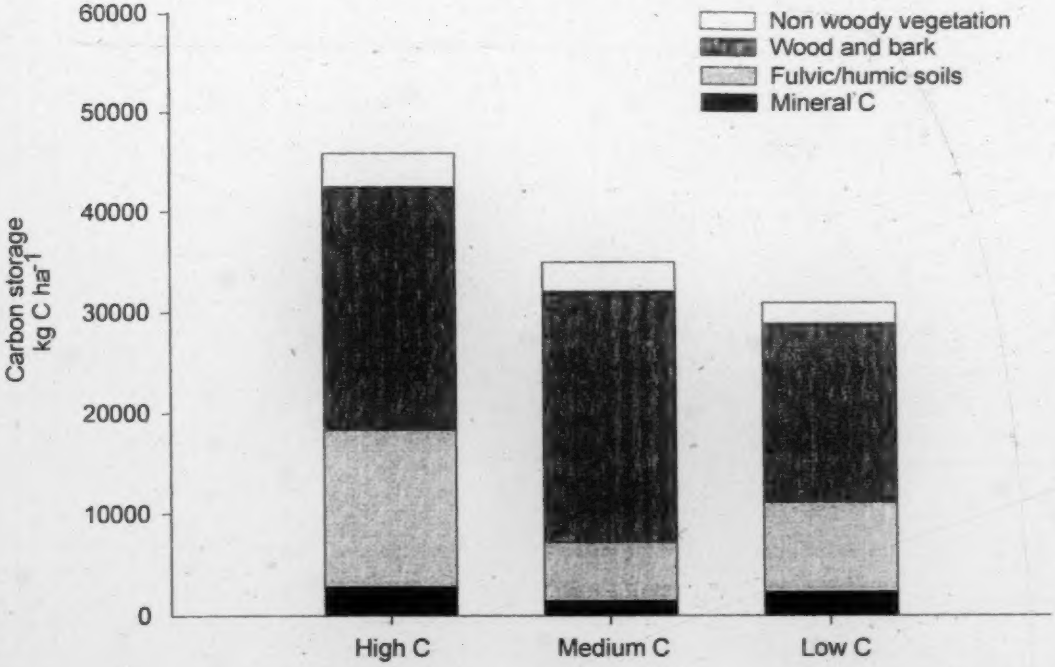

Figure 3-1. Carbon stores (kg C ha^{-1}) in the FLUDEX reservoirs prior to flooding.

While initial rates of decomposition were slower in tissues placed in unflooded forests compared to the same tissues placed in reservoirs, there were no differences in the amount of C and N mass left in the tissues at the end of the study among sites. Depending on tissue type, decomposing litter in both natural forests and reservoirs was either a source or a sink for THg. In tissues that lost THg, the mass of THg declined more rapidly in plant tissues placed in reservoirs compared to the same plant tissue placed in unflooded forests, but there were no differences in final THg masses after 800 days. Plant tissues placed in flooded forests exhibited large increases in MeHg mass, whereas MeHg mass decreased in the same plants placed in unflooded forests. This is the first study examining THg and MeHg cycling in decomposing plants in upland boreal forests.

Methods

Litterbag construction

Twelve plant tissues were used in this study (Table 3-1). Six plant tissues were common to all three upland sites (birch [*Betula papyrifera*], alder [*Alnus crispa*], and blueberry [*Vaccinium myrtilloides* and *V. angustifolium.*] leaves, bunchberry [*Cornus canadensis*] plants, jack pine [*Pinus banksiana*] needles, and wood). The other plants were specific to individual sites (Labrador tea leaves [*Ledum groenlandicum*], mosses [*Sphagnum* spp., *Polytrichum* spp., and *Pleurozium* spp.], lichen [*Cladina* spp.], and old wood; Table 3-1). In the spring of 1999, birch, alder, blueberry, and Labrador tea leaves and jack pine needles were picked from trees and shrubs. Vinyl gloves were worn when handling all material and plant tissues were stored in clean plastic Ziploc bags. Bunchberry samples consisted of both the aboveground leaves and stems. *Sphagnum* spp. were collected from wetland patches in the upland forests and the capitulum (growing tip) was removed from each plant to ensure there was no re-growth *in situ*. Upland dwelling mosses (*Pleurozium* spp. and *Polytrichum* spp.) were collected from large patches of moss located in a near-by old growth forest and trimmed to exclude the rhizoids. Lichen (*Cladina* spp.) was collected from exposed bedrock on ridge top and trimmed to exclude soil.

All samples were dried at ambient temperature for two weeks on labmat in a clean laboratory at the ELA to avoid contamination of plant samples. Approximately 5 g dry weight (d.w.) of each plant tissue was placed in a 10 x 10 cm litterbag constructed of acid washed 400 μm Nitex mesh sewn together using heavy nylon thread. Litterbags were also filled with wooden blocks (~13 g) cut from a single piece of fir lumber containing no sapwood and knots and ~5 g pieces of extensively decomposed wood ('old wood') collected from fallen decomposing trees near the reservoir sites.

Table 3-1. Types of plant tissues placed in litterbags in unflooded forests and reservoirs.

Plant tissue	High C unflooded forests and reservoirs	Medium C unflooded forests and reservoirs	Low C unflooded forests and reservoirs
Living trees			
Birch leaves (*Betula papyrifera*)	✓	✓	✓
Pine needles (*Pinus banksiana*)	✓	✓	✓
Wood blocks (*Abies* spp.)	✓	✓	✓
Herbs and shrubs			
Alder leaves (*Alnus crispa*)	✓	✓	✓
Blueberry leaves (*Vaccinium* spp.)	✓	✓	✓
Bunchberry plants (*Cornus canadensis*)	✓	✓	✓
Labrador tea leaves (*Ledum groenlandicum*)	✓		
Bryophytes			
Sphagnum spp.	✓		
Polytrichum spp.			✓
Pleurozium spp.			✓
Lichen (*Cladina* spp.)			✓
Old wood		✓	

Litterbag placement and site descriptions

The ELA is a series of Boreal lakes set aside by the Canadian Federal and Ontario Provincial governments for limnological and ecological research. The ELA camp is located 50 km southeast of Kenora, Ontario, on the Precambrian Shield (Brunskill and Schindler 1971). The ELA experiences a cold temperate continental climate with 32-year mean July and January temperatures of 18.5 and –17.3°C, respectively. Mean annual wet deposition from 1969-2001 was 699 mm, with ~25% of this wet deposition falling as snow. Upland areas at the ELA ranged from open lichen-covered granite/gneiss rocks to shallow nutrient-poor acidic soils supporting jack pine (*Pinus banksiana*), black spruce

(*Picea mariana*), and paper birch (*Betula papyrifera*) forest communities.

To compare decomposition rates and changes in THg and MeHg content in plants in unflooded and flooded forests, five sets of triplicate litterbags containing each plant tissue type were placed on top of existing litter in three unflooded forests (Low C, Medium C, and High C unflooded sites) and in three reservoirs (Low C, Medium C, and High C reservoirs). We also wanted to compare rates of decomposition at different depths in the reservoirs, so five sets of triplicate litterbags were placed in shallow (<25 cm) and deep (>1 m) zones of each reservoir.

Our forest and reservoir sites differed in aerial coverage and species composition of plant communities, so litterbags placed in each site contained plant tissues characteristic of the site. The High C site was a wet jack pine dominated forest, with an understory of wetland plants. Litterbags containing the six common plant tissues (birch, alder, and blueberry leaves, bunchberry plants, jack pine needles, and wood) and *Sphagnum* spp. moss and Labrador tea leaves were placed in a hollow in the wet jack pine dominated forest community outside of the flooded area and at two locations in the 0.74 ha High C reservoir (Fig. 3-2). The Medium C site was a dense jack pine forest with birch and alder, with understory of blueberry shrubs and various mosses and herbs. Litterbags containing the six common plant tissues and old wood were placed in this homogeneous forest, as well as in two locations in the 0.50 ha Medium C reservoir (Fig. 3-3). The Low C site had shallow soils supporting sparse stands of jack pine and birch with a blueberry shrub dominated understory, and areas of thin glacial till with lichens, mosses, blueberry shrubs, and exposed bedrock. Litterbags containing the six common tissues, as well as mosses (*Pleurozium* spp. and *Polytrichum* spp.) and lichen, were placed in the jack pine, birch, and blueberry dominated forest stand and in two locations in the 0.63 ha Low C reservoir (Fig. 3-4).

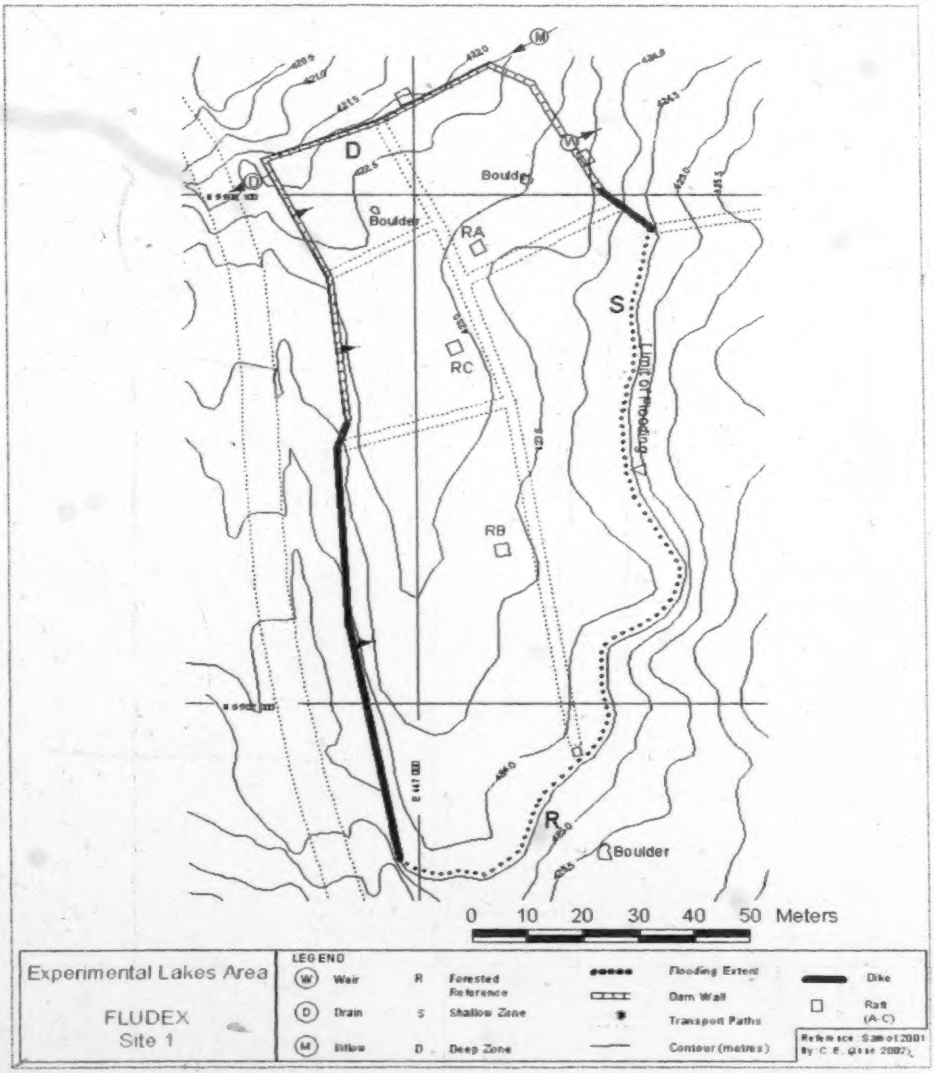
D
Boulder
Boulder
RA
RC
RB
S
Limit of flooding
R
Boulder
0 10 20 30 40 50 Meters
Experimental Lakes Area
FLUDEX
Site 1
LEGEND
W Weir
D Drain
M Inflow
R Forested Reference
S Shallow Zone
D Deep Zone
Flooding Extent
Dam Wall
Transport Paths
Contour (metres)
Dike
Raft (A-C)
Reference: Sam et 2001
By: C. E. Gase 2002

Figure 3-2. Location of litterbags in the High C site. Litterbag locations are indicated by the R (unflooded forests), S (shallow), and D (deep).

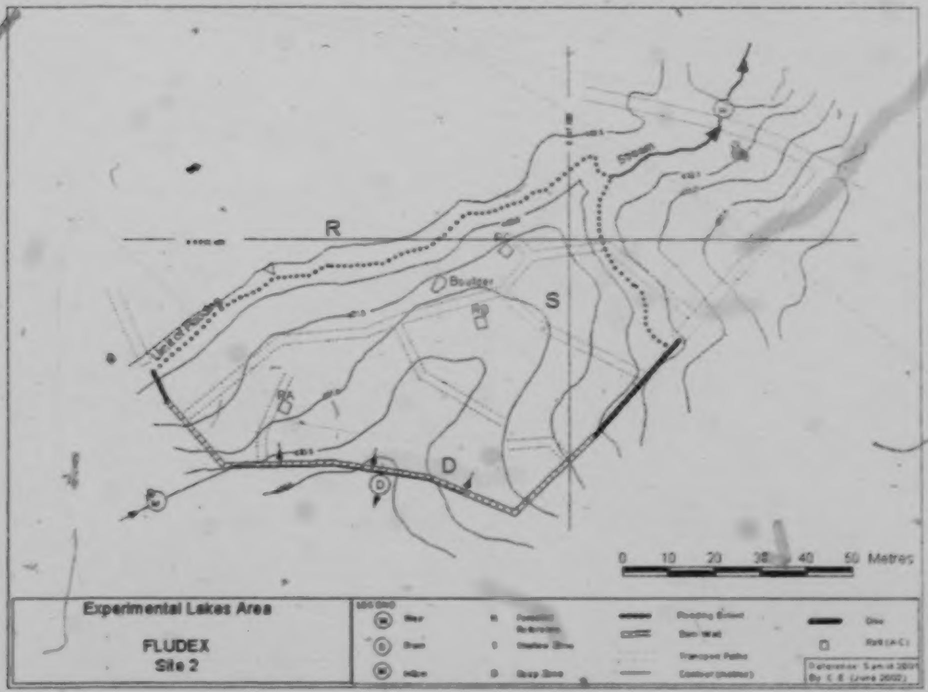

Figure 3-3. Location of litterbags in the Medium C site. Litterbag locations are indicated by the R (unflooded forests), S (shallow), and D (deep).

Litterbag collection and processing

Three litterbags of each plant tissue type were retrieved from unflooded and flooded sites in autumn 1999 (the first summer that the reservoirs were flooded), and in spring and autumn 2000 and 2001. Any foreign debris on the outside of each litterbag was removed by gloved hand. Litterbags were then allowed to dry on labmat map in a clean laboratory at the ELA at ambient temperature for two weeks. Once dry, plant tissues were removed from the litterbags, weighed, and freeze-dried. Freeze-dried tissues from each litterbag were ground using an acid-cleaned coffee grinder, with the exception of wood blocks, which were shaved using a clean stainless steel rasp. A portion of each dried sample was analyzed for concentrations of C, N, MeHg and THg. Large amounts of plant tissue were retained at the beginning of the study to measure initial concentrations.

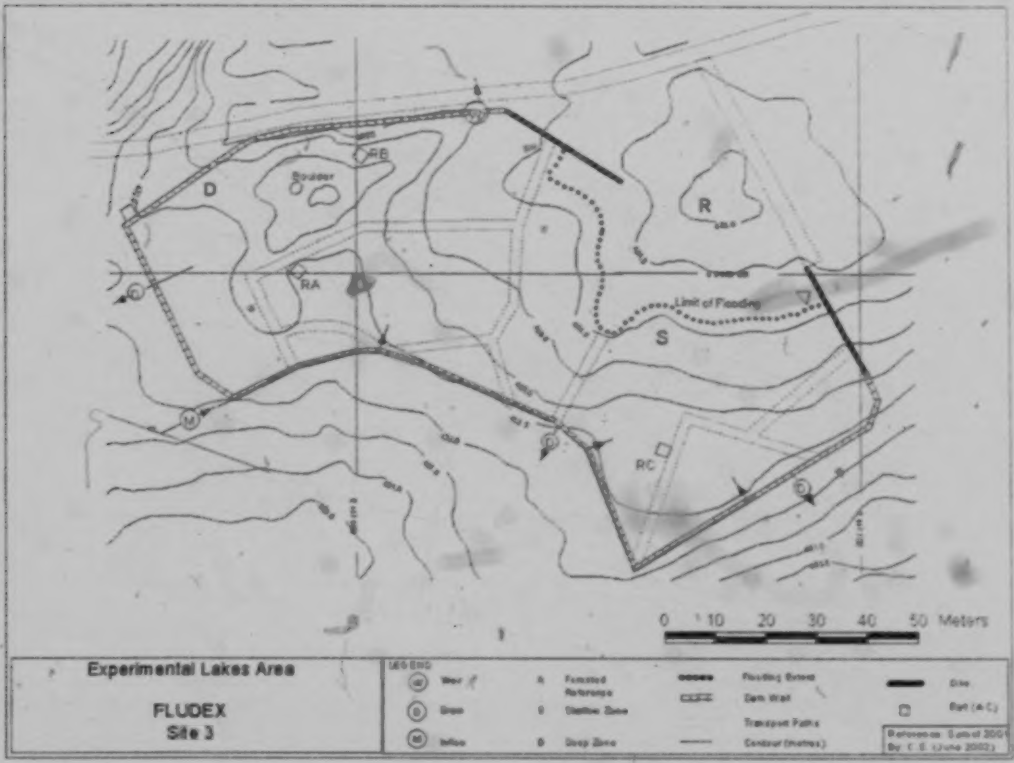
D
Boulder
RB
RA
R
Limit of Flooding
S
RC
0 10 20 30 40 50 Meters
Experimental Lakes Area
FLUDEX
Site 3
Forested Reference
Shallow Zone
Deep Zone
Transport Paths
Contour (metres)

Figure 3-4. Location of litterbags in the Low C site. Litterbag locations are indicated by the R (unflooded forests), S (shallow), and D (deep).

Analytical methods

Carbon and nitrogen

C and N content of plant tissues were analyzed using an Exeter Analytical Model 440 elemental analyser at the University of Alberta Limnology Laboratory (Edmonton, Alberta) and a Carlo Erba EA1108 elemental analyzer at the University of Waterloo Environmental Geochemistry Laboratoryy (Waterloo, Ontario). The mass of C and N in each litterbag was calculated by multiplying the C or N content (%C or %N) by the final air-dried weight of tissue in the litterbag. The percent mass of C or N remaining in each litterbag was standardized to the initial C or N mass placed into each litterbag and presented as an average ± one standard error. C:N ratios were also calculated for each sample.

We tested for differences in C and N mass and C:N ratios in plants placed among deep, shallow, and unflooded sites within the High C, Medium C, and Low C sites using ANOVA (Minitab Version 9.2, 1993) (Wieder and Lang 1982). We were unable to test C and N mass differences among the different sites, however, due to lack to replication in whole ecosystem experiments.

Mercury

All Hg analysis was performed in the University of Alberta Low-level Hg Analytical Laboratory. For THg and MeHg analyses, equal amounts of freeze dried tissue from the three triplicate litterbags were pooled together and analyzed to provide one THg or MeHg concentration per tissue per sampling date. Due to the high cost associated with Hg analysis, THg was only analyzed for one tissue from the deep, shallow, and unflooded sites on three out of five sampling dates (each autumn sampling date). MeHg was only analyzed on six tissues (jack pine needles, blueberry and Labrador tea leaves, *Pleurozium* spp., *Polytrichum* spp., and lichen) from the forests and the deep station in the three reservoirs, for two out of five sampling dates (autumn, years 1 and 3).

THg analyses were performed on 50-200 mg of ground, freeze-dried plant tissue digested in 7 mL of 7:3 (vol:vol) HNO_3:H_2SO_4 in sealed Teflon digestion bombs. Samples were initially digested at 125°C for two hours, after which 1 mL BrCl and 19 mLs of distilled, deionized water were added. Samples were then heated overnight at 60°C. 50μL-3mL of digested sample was then placed into glass bubblers with 1 mL $SnCl_2$ (to reduce all HgII to Hg^0) and 200 μL of $NH_2OH*HCl$ (to neutralize the oxidation to HgII). Samples were purged for 10 minutes using ultra high purity (UHP) N_2. During the purging process, Hg^0 was transferred to traps containing gold-coated glass beads (gold traps) and dried in the N_2 stream for 10 minutes. Mercury on gold traps was then thermally desorbed to the analytical system and THg was detected using cold vapour atomic fluorescence spectrophotometry (CVAFS) as described in Bloom and Fitzgerald (1988) with detection at 0.1-0.3 ng g^{-1} at a blank level of 0.3-0.4 ng L^{-1}. Standard reference material (NRC-TORT) and standards were analyzed in tandem with all samples and concentrations were always within 10% of certified values. Spike recoveries were always within 90-110% of original spike addition.

For MeHg analysis, ~300 mg of processed plant tissues were digested overnight at 60°C in Teflon bottles containing 5 mL of 25% KOH in methanol. After digestion, a 1mL aliquot of the digest was centrifuged and 500-800 µL of supernatant were distilled at 135 °C in 2.5 mLs of 2:1:2 (vol:vol:vol) H_2SO_4:KCl:$CuSO_4$ until 85% of the sample was transferred to the receiving jar. The distillate was then placed in glass bubblers with 500 µL acetate buffer and 100 µL Na tetraethyl borate and purged onto Carbotraps using UHP N_2. Mercury species were thermally desorbed from Carbotraps, separated using a gas chromatography column, reduced using a pyrolytic column, and detected by CVAFS (Bloom 1989; Horvat and others 1993; Olson and others 1997) (detection limits = 0.1-0.3 ng g^{-1} at a blank level of 0.05 ng L^{-1}). Spike recovery for MeHg analysis was between 80% and 120% of original added spike and concentrations in standard reference material (NRC-TORT) and analytical standards were always within 10% of certified values. 10% of THg and MeHg samples were analyzed in duplicate.

The mass of THg and MeHg in each litterbag was calculated by multiplying the THg or MeHg concentration by the final air-dried weight of tissue in the litterbags, giving us three THG masses for each tissue, from each site, on each date. The percent mass of THg or MeHg remaining in each litterbag was standardized to the initial THg or MeHg mass placed into each litterbag and data are presented as an average ± one standard error. Statistical analysis could not be performed on THg and MeHg data because plant tissue was pooled prior to analysis.

Results

Initial C and N concentrations and C:N ratios in plant tissue

Initial C content in birch, blueberry, alder, and Labrador tea leaves, jack pine needles, and wood was between 49.8±0.04 %C and 53.3±0.3 %C (Table 3-2). Percent C in bunchberry plants, bryophytes (*Sphagnum* spp., *Pleurozium* spp., and *Polytrichum* spp.), and lichens had slightly lower %C (45.8±0.02 %C - 48.3±0.2 %C). The highest C content was in old wood chunks (59.6±0.11 %C; Table 3-2).

Initial N content ranged from 0.03±0.01 %N in old wood to 2.83±0.12 %N in alder leaves (Table 3-2). Lichens, wood, and old wood had the lowest N content (0.03 %N - 0.2 %N), jack pine needles, Labrador tea leaves, and bryophytes had intermediate levels (1.0 %N - 1.5 %N) and birch, blueberry, and alder leaves and bunchberry plants had the highest N content (2.3 %N – 2.8 %). C:N ratios were lowest in birch, blueberry, and alder leaves and bunchberry plants (18.4 - 22.3; Table 3-2) and highest in old wood and wood (199 and 226, respectively). Labrador tea leaves and jack pine needles had intermediate C:N ratios (33.5 and 41.0, respectively). In bryophytes, the C:N ratios ranged from 31.0 (*Polytrichum* spp.) to 46.6 (*Sphagnum* spp.) and lichens exhibited a C:N ratio of 76.3 (Table 3-2).

Initial MeHg and THg concentrations in plant tissue

Initial THg concentrations in plant tissue ranged from under 5 ng g^{-1} to over 90 ng g^{-1} (Table 3-2). THg concentrations were lowest in wood (3.62 ng g^{-1}), birch and blueberry leaves (7.13 and 5.75 ng g^{-1}, respectively), bunchberry plants (9.77 ng g^{-1}), and old wood (9.68 ng g^{-1}). Moderate THg concentrations were

Table 3-2. Tissue mass, carbon and nitrogen content, C:N ratios, and total mercury (THg) and methylmercury (MeHg) concentrations in tissues in litterbags prior to placement in natural forests and reservoirs. nd= no data.

Plant tissue	Tissue mass (g litterbag^{-1})	C content (%C)	N content (%N)	C:N	THg (ng g^{-1})	MeHg (ng g^{-1})
Living trees						
Birch leaves	5.12±0.02	49.8±0.04	2.4±0.05	21.1	7.13	nd
Pine needles	5.16±0.01	52.1±0.1	1.3±0.01	41.0	14.07	0.088
Wood blocks	12.81±0.04	52.1±0.4	0.2±0.01	226.4	3.62	nd
Herbs and shrubs						
Alder leaves	5.11±0.01	52.1±0.1	2.8±0.12	18.4	12.84	nd
Blueberry leaves	5.06±0.004	51.4±0.1	2.3±0.02	22.3	5.75	0.185
Bunchberry plants	5.10±0.01	46.1±0.4	2.4±0.02	19.1	9.77	nd
Labrador tea leaves	5.37±0.03	53.3±0.3	1.6±0.03	33.5	27.13	0.447
Bryophytes						
Sphagnum spp.	5.07±0.01	46.6±0.3	1.0±0	46.6	52.57	nd
Polytrichum spp.	5.10±0.01	47.5±0.3	1.5±0.07	31.0	93.85	0.398
Pleurozium spp.	5.01±0.01	48.4±0.2	1.0±0.02	46.5	64.64	1.232
Lichen	5.15±0.01	45.8±0.02	0.1±0.01	76.3	33.17	0.556
Old wood	5.17±0.04	59.6±0.1	0.03±0.01	198.6	9.68	nd

found in Labrador tea leaves (27.13 ng g^{-1}) and lichens (33.17 ng g^{-1}). Bryophytes had the highest THg concentrations (52.57 - 93.85 ng g^{-1}).

The lowest initial MeHg concentrations were found in blueberry leaves (0.185 ng g^{-1}) and jack pine needles (0.088 ng g^{-1}; Table 3-2). Intermediate MeHg concentrations were found in Labrador tea leaves, *Polytrichum* spp., and lichens (0.447, 0.398, 0.556 ng g^{-1}, respectively). The feather moss, *Pleurozium* spp., had the highest initial MeHg concentrations (1.232 ng g^{-1}; Table 3-2).

Decomposition of litter in unflooded and flooded forests

Change in tissue mass

- Unflooded forests

Birch, alder, blueberry, and Labrador tea leaves and bunchberry plants placed in unflooded forests lost between 20% and 55% of their original tissue mass during the first summer of study (Figs. 3-5 to 3-9). Jack pine needles, bryophytes, and lichens initially lost between <5% to 15% of their original mass (Figs. 3-10 to 3-14). Loss of mass from all plant tissues placed at unflooded sites, with the exception of wood and old wood, was highest during the first year, but continued to decline at gradual and constant rates in subsequent years, with final mass losses of 25% - 80% of original mass after 800 days. Wood and old wood exhibited very little to no mass loss during the study period (Figs. 3-15, 3-16). For each type of plant tissue, there were generally no differences in mass loss among the three unflooded forest sites (Appendix 2).

- Reservoirs

Mass loss in leaves and bunchberry plants in reservoirs was highest during the first summer of flooding (30% - 80% of original mass lost), but levelled off until the final summer of the study when a few tissues showed further decreases in mass (Figs. 3-5 to 3-9).

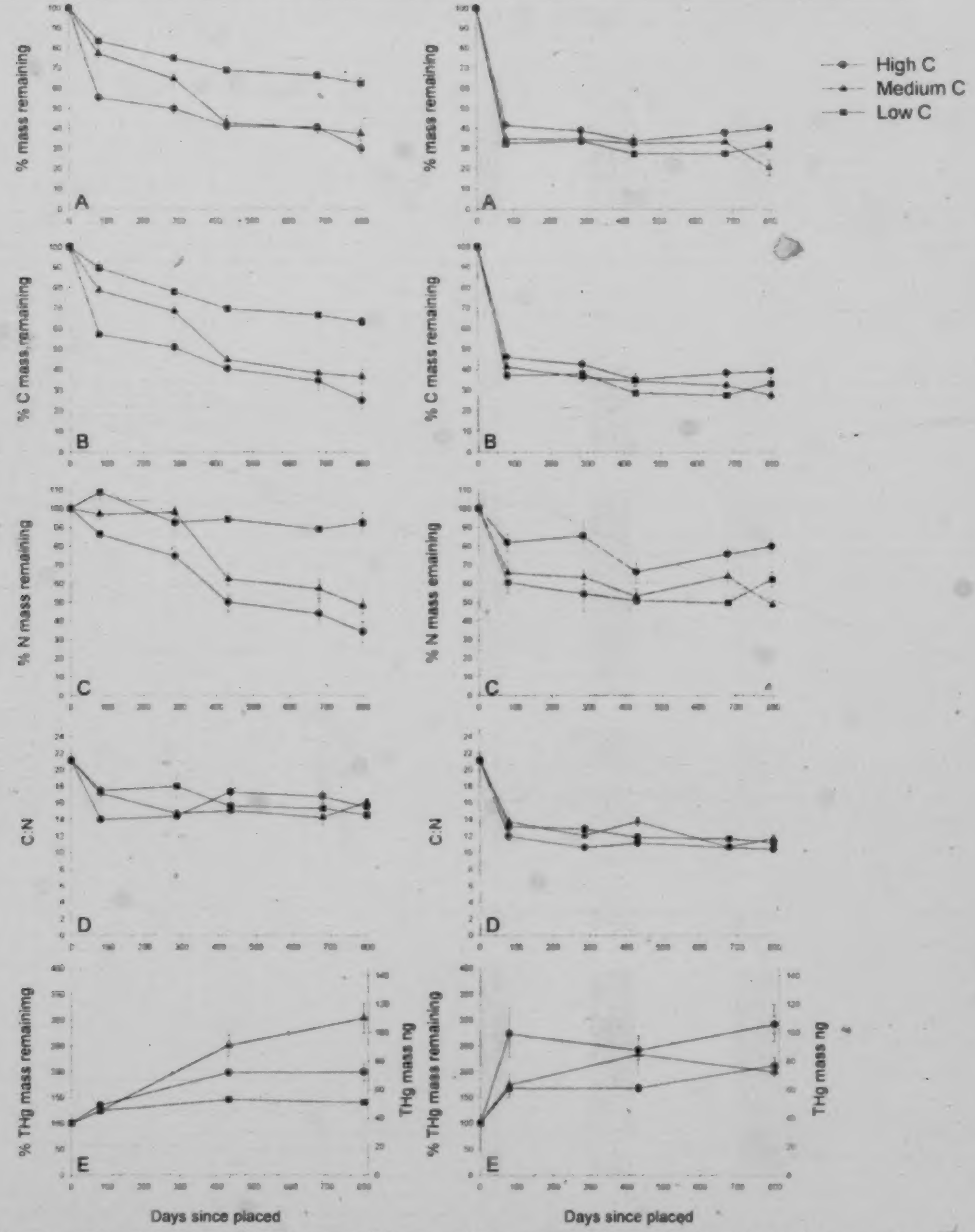

Figure 3-5. Rates of decomposition and mercury changes in birch leaves in unflooded and flooded forests. A: Percent of original tissue mass remaining. B: Percent of original C mass remaining. C: Percent of original N mass remaining. D: C/N ratios. E: Total mass and the percent of original total mercury remaining. Shaded areas represent periods of inundation.

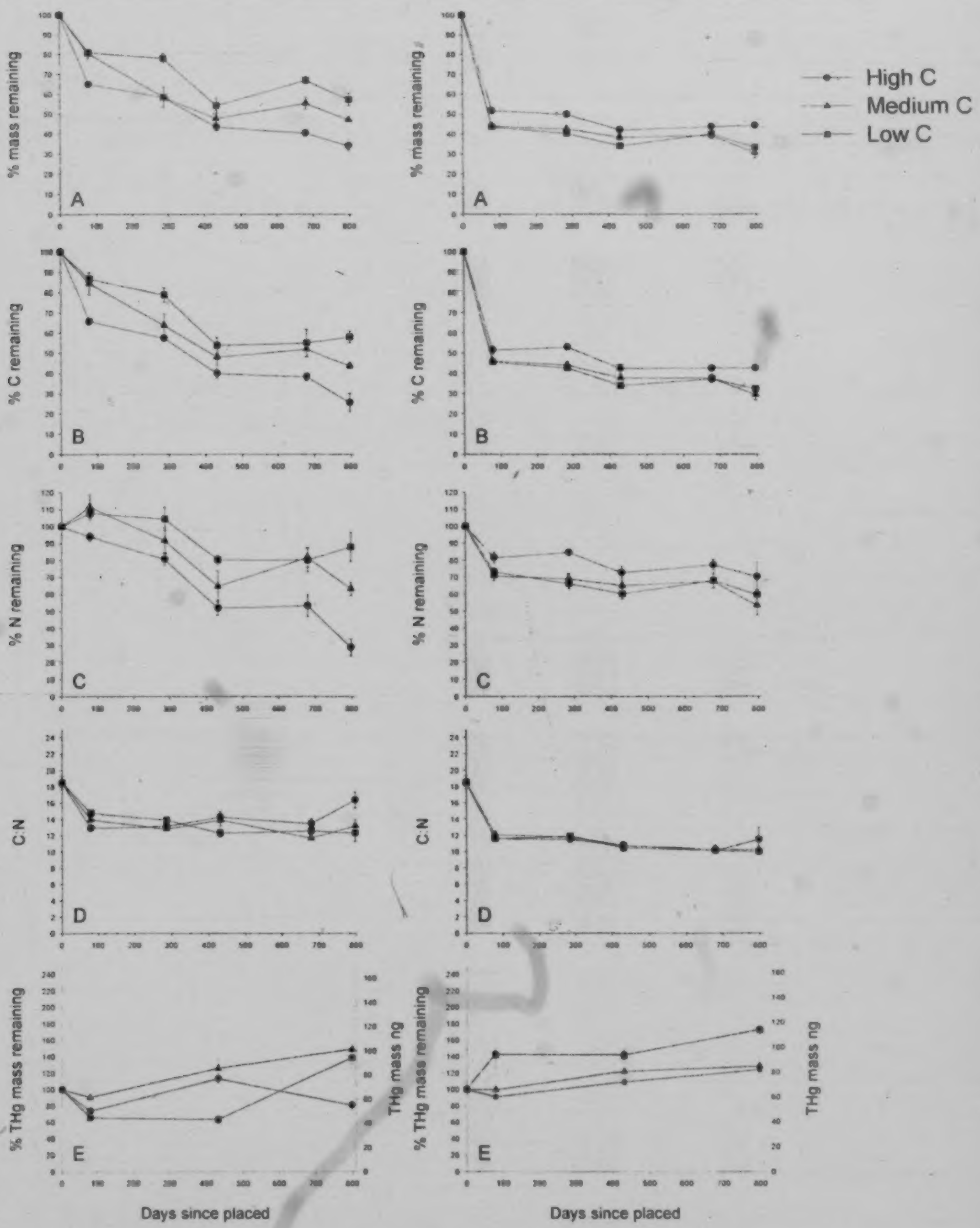

Figure 3-6. Rates of decomposition and mercury changes in alder leaves in unflooded and flooded forests. For explanation of figure, see Figure 3-5.

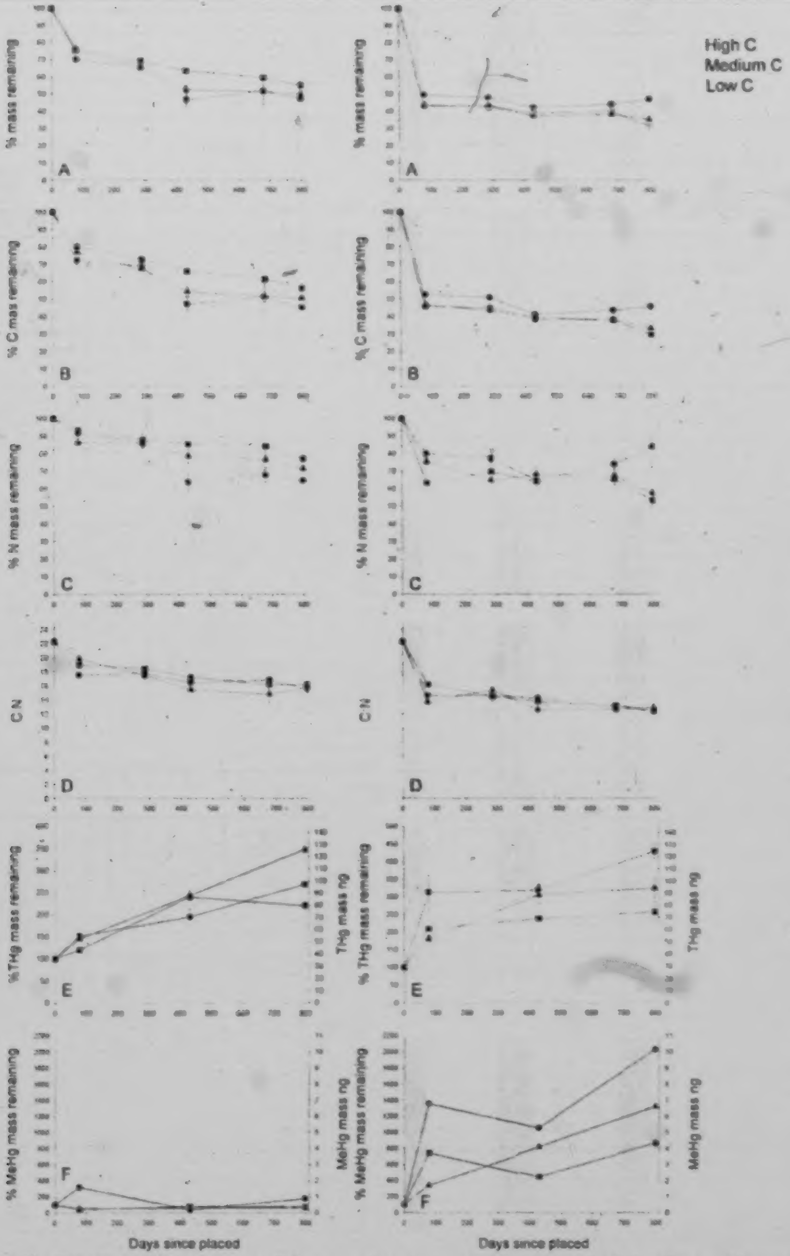

Figure 3-7. Rates of decomposition and mercury changes in blueberry leaves in unflooded and flooded forests. A: Percent of original tissue mass remaining. B: Percent of original C mass remaining. C: Percent of original N mass remaining. D: C/N ratios. E: Total mass and the percent of original total mercury remaining. F: Total mass and the percent of original methylmercury remaining. Shaded areas represent periods of inundation.

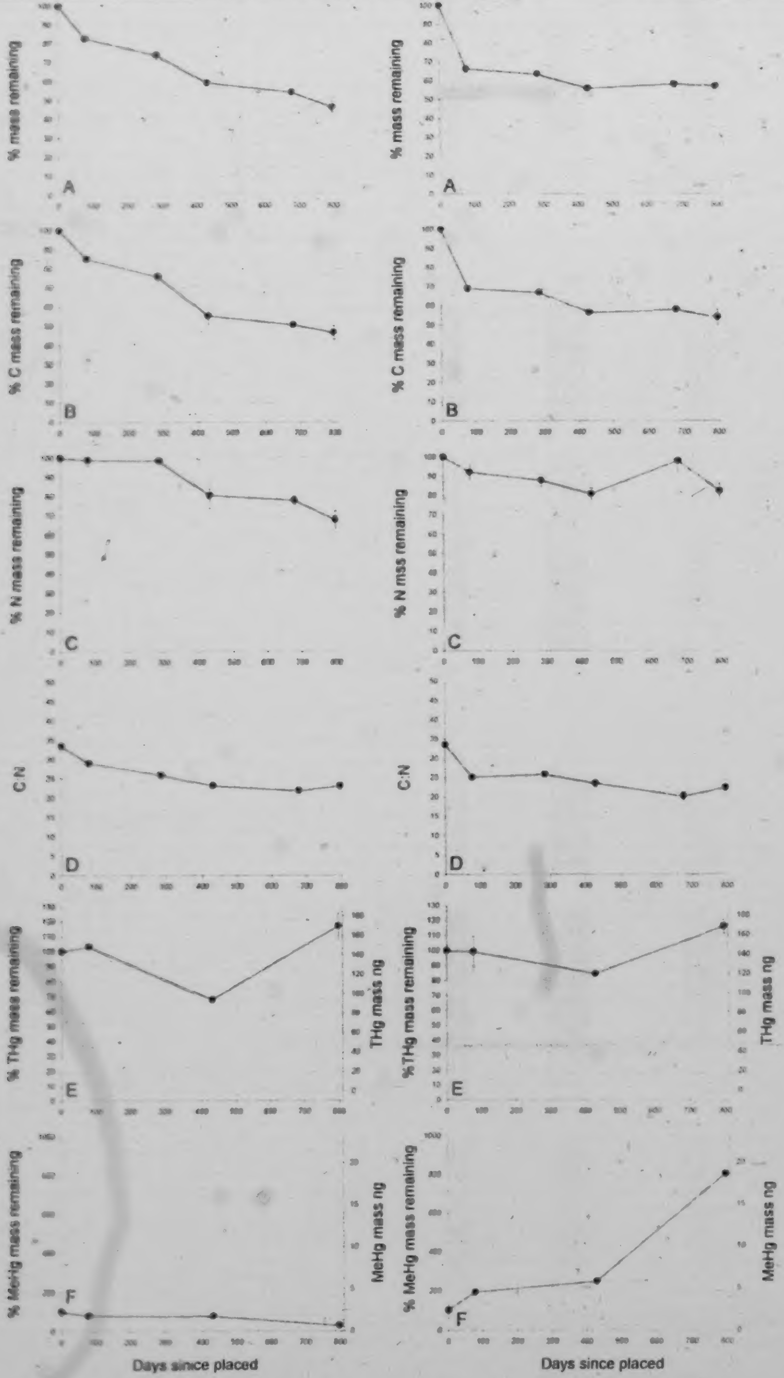

Figure 3-8. Rates of decomposition and mercury changes in Labrador tea leaves in unflooded and flooded forests. For explanation of figure, see Figure 3-7.

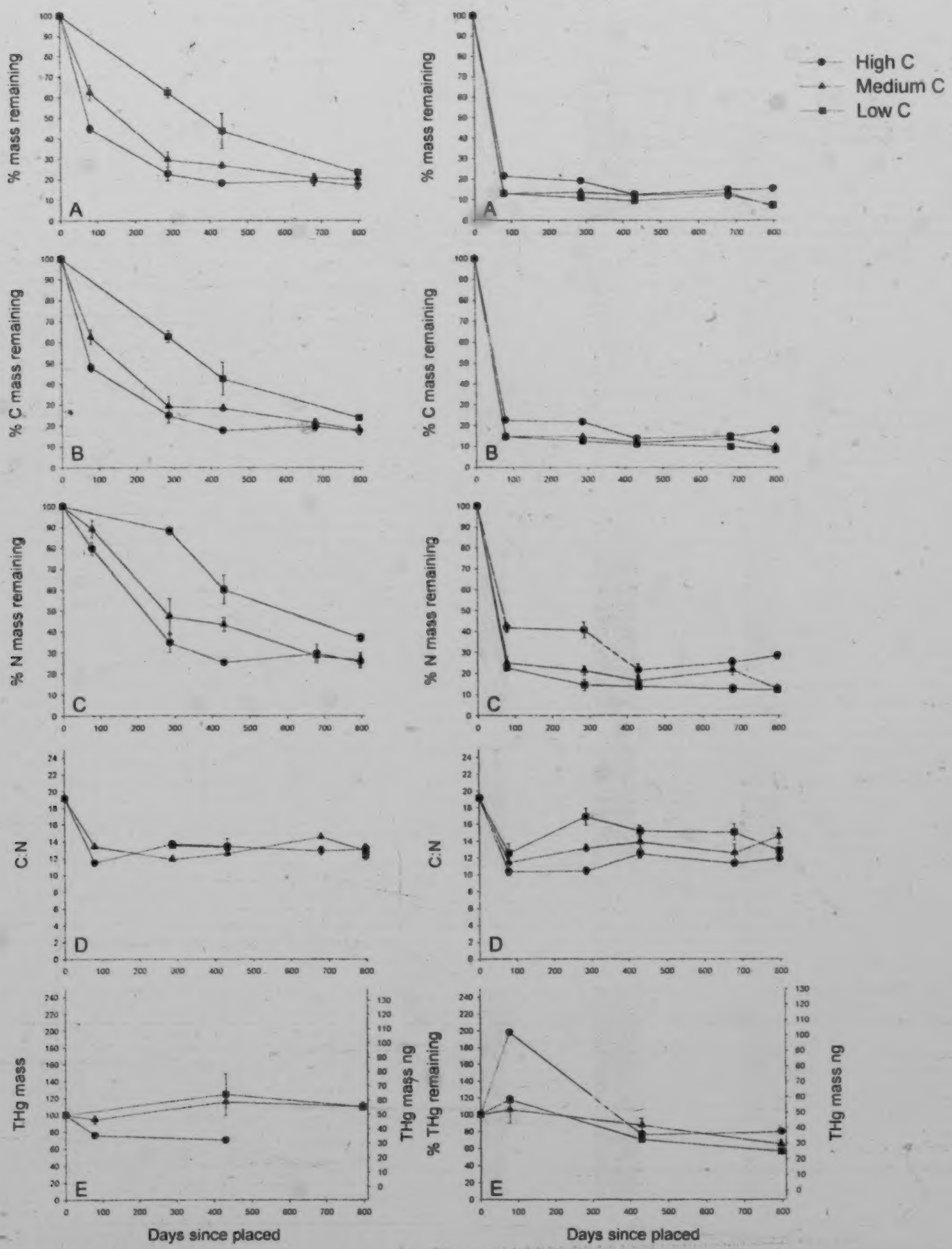

Figure 3-9. Rates of decomposition and mercury changes in bunchberry plants in unflooded and flooded forests. For explanation of figure, see Figure 3-5.

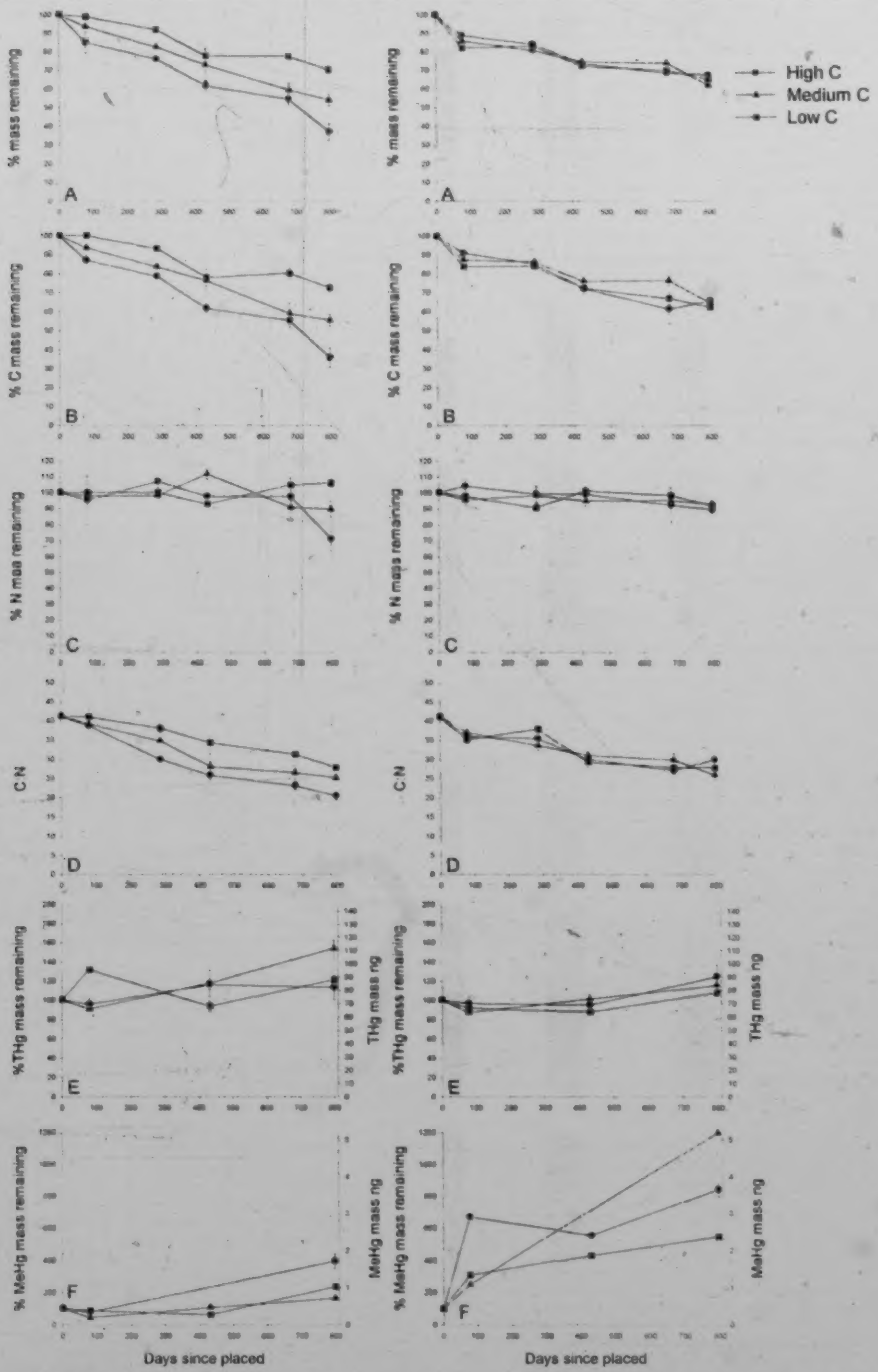

Figure 3-10. Rates of decomposition and mercury changes in jack pine needles in unflooded and flooded forests. For explanation of figure, see Figure 3-7.

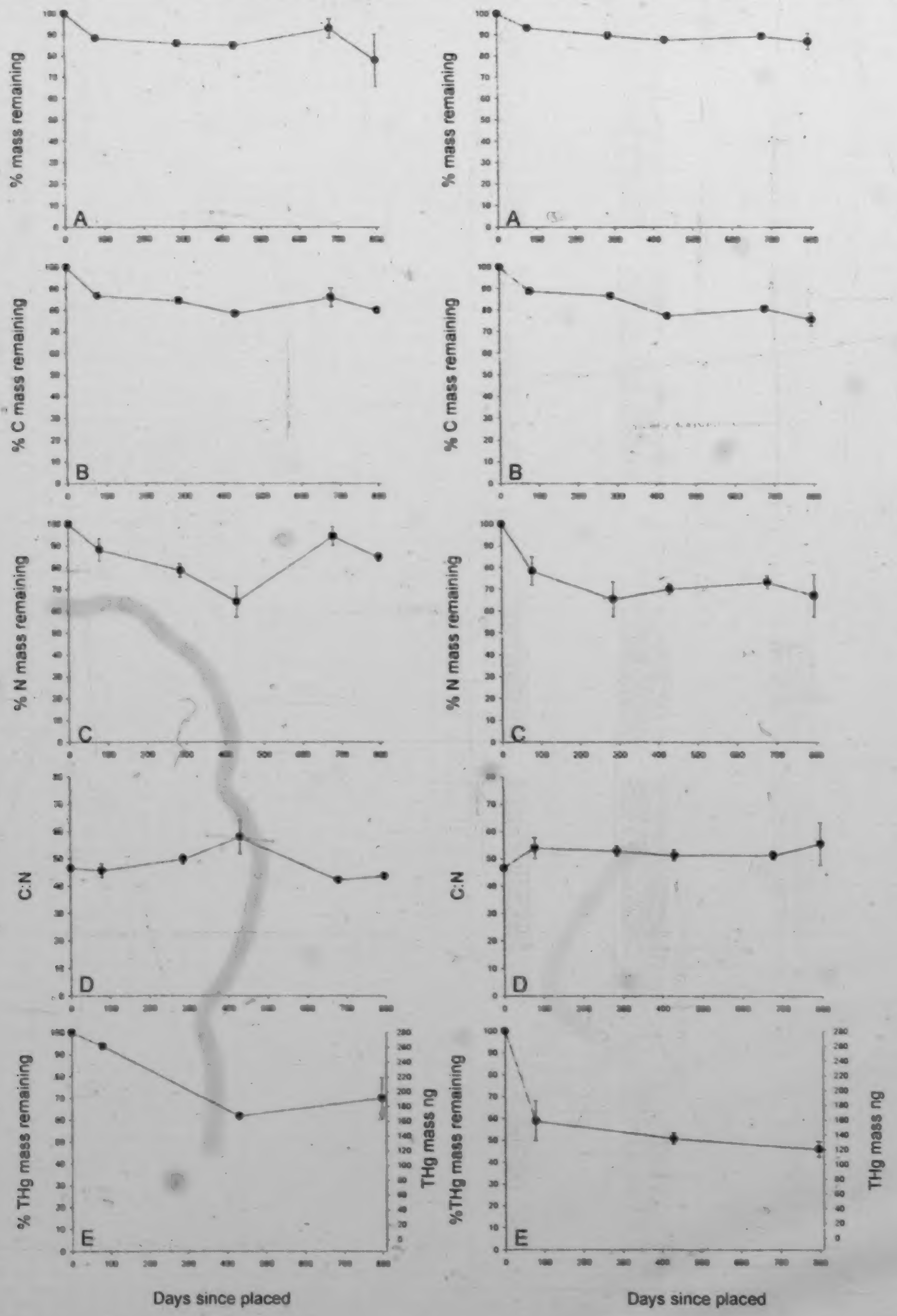

Figure 3-11. Rates of decomposition and mercury changes in *Sphagnum* spp. in unflooded and flooded forests. For explanation of figure, see Figure 3-5.

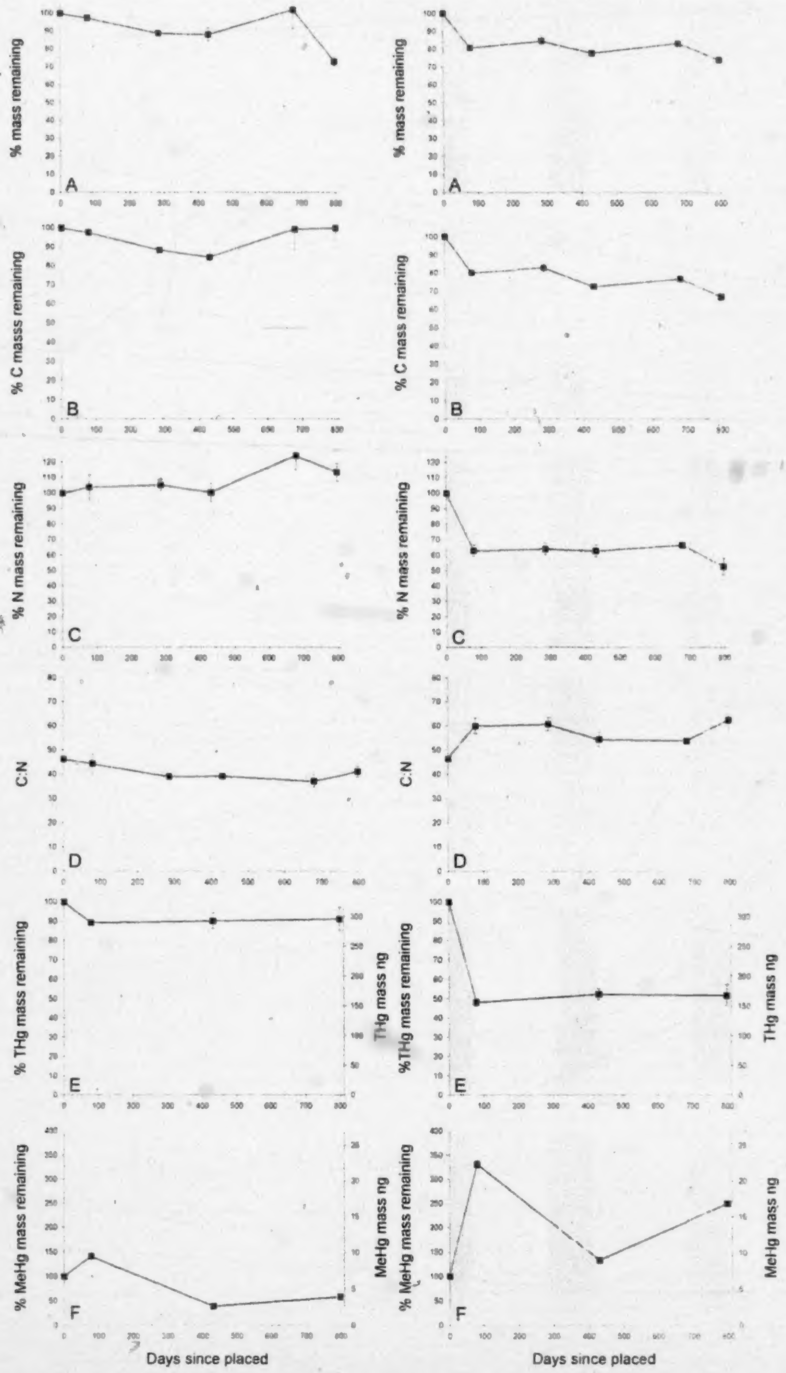

Figure 3-12. Rates of decomposition and mercury changes in *Pleurozium* spp. in unflooded and flooded forests. For explanation of figure, see Figure 3-7.

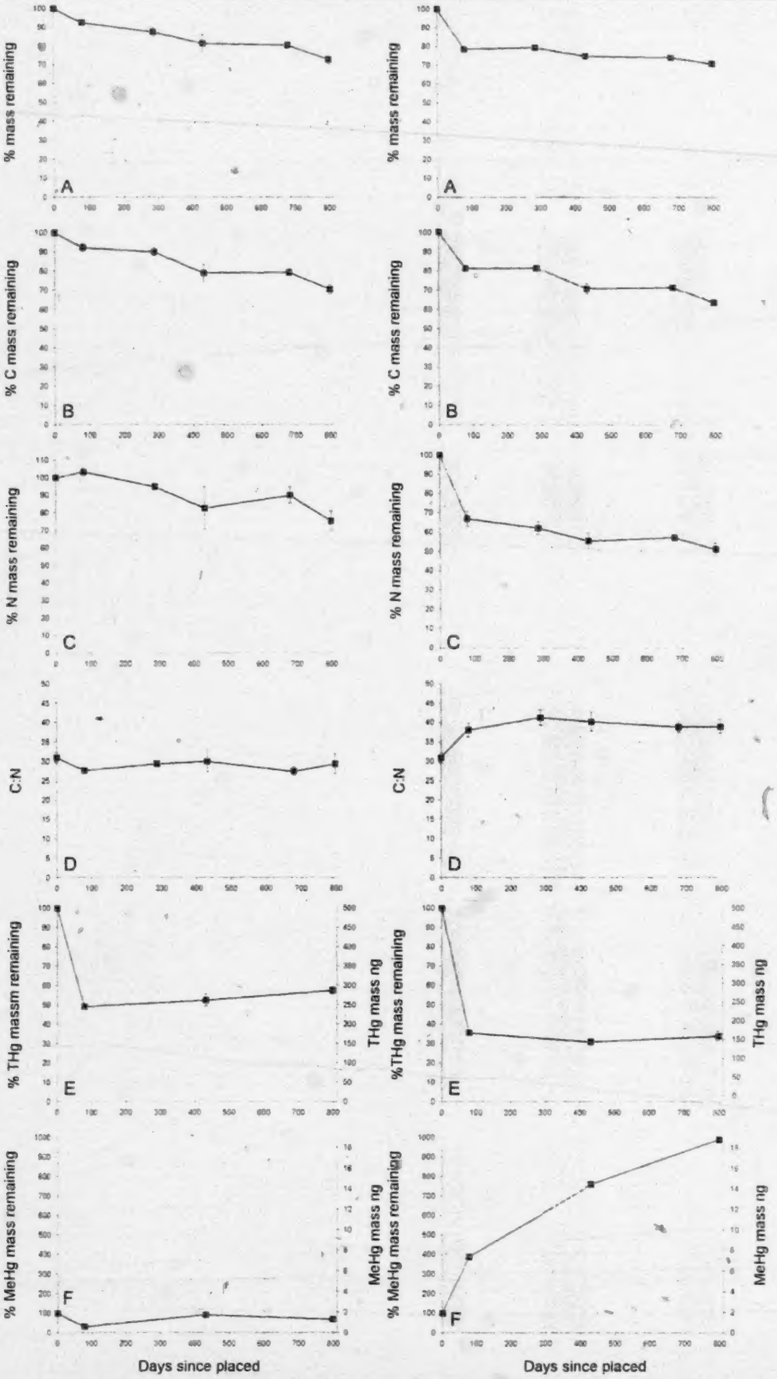

Figure 3-13. Rates of decomposition and mercury changes in *Polytrichum* spp. in unflooded and flooded forests. For explanation of figure, see Figure 3-7.

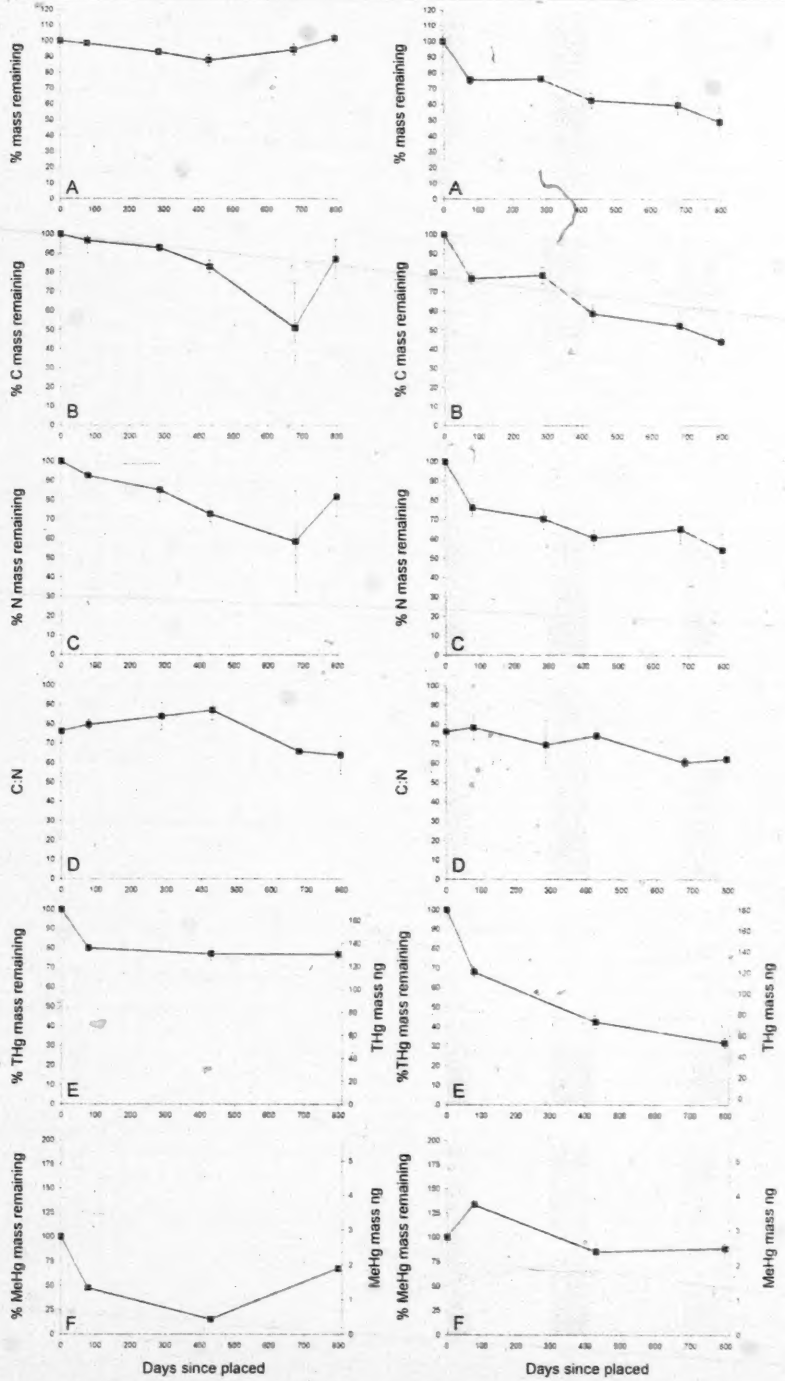

Figure 3-14. Rates of decomposition and mercury changes in lichen in unflooded and flooded forests. For explanation of figure, see Figure 3-7.

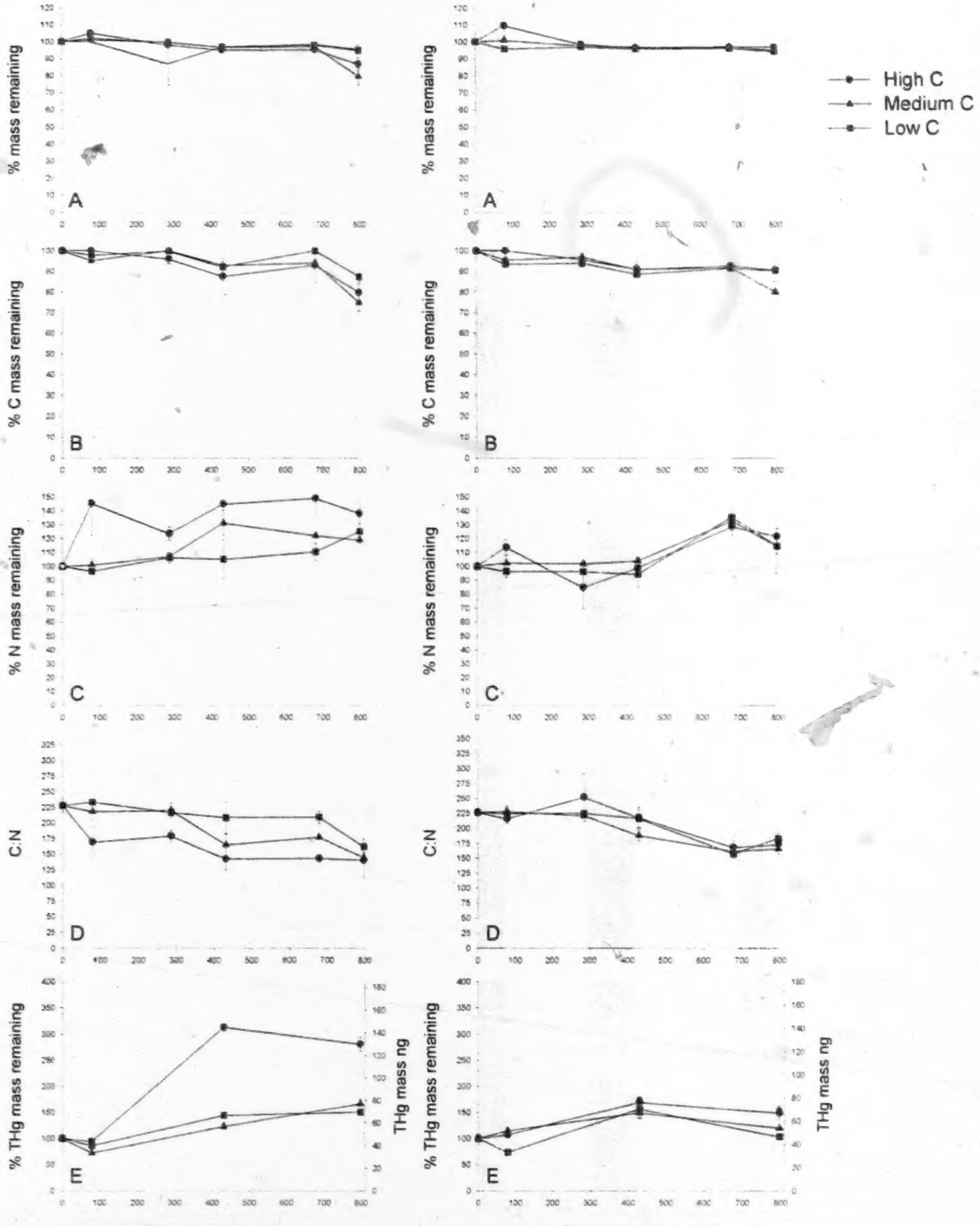

Figure 3-15. Rates of decomposition and mercury changes in wood in unflooded and flooded forests. For explanation of figure, see Figure 3-5.

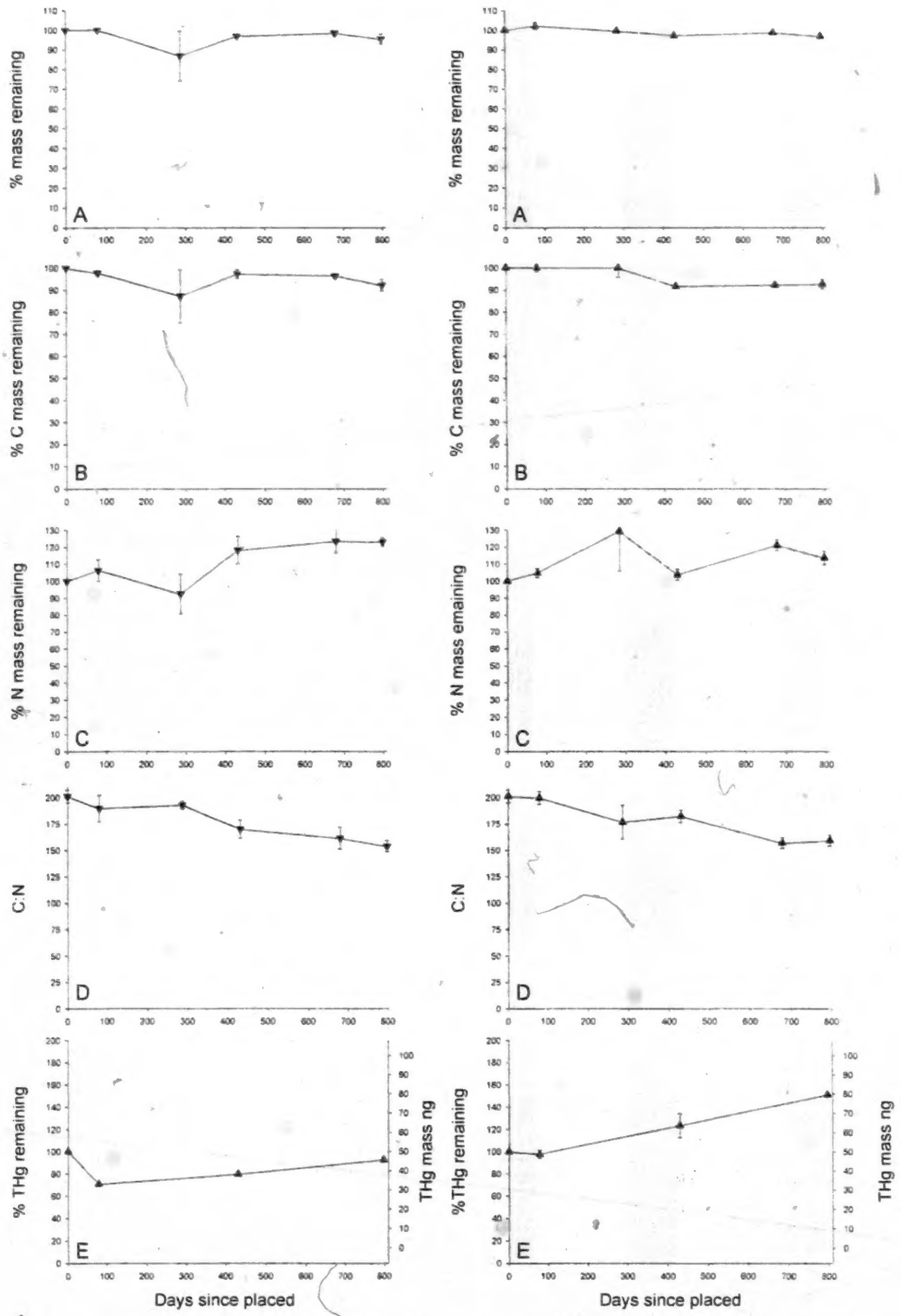

Figure 3-16. Rates of decomposition and mercury changes in old wood in unflooded and flooded forests. For explanation of figure, see Figure 3-5.

Flooded jack pine needles, bryophytes, and lichens initially lost between 10% and 30% of their original mass (Figs. 3-10 to 3-14). Mass of jack pine needles (Fig. 3-10) and lichens (Fig. 3-14) constantly declined over the entire ~800 day study period, with final mass losses of 30%-50% of original mass. Bryophytes, however, only lost a maximum of 30% of their mass by the end of the three-year study (Figs. 3-11 to 3-13). Wood and old wood exhibited very little to no mass loss during the study period (Figs. 3-15 to 3-16). For each of the six tissues found in all three reservoirs, there were generally no differences in the rates of mass loss within or among reservoirs or depths (Figs. 3-5 to 3-9, 3-11 to 3-14).

All plant tissues placed in reservoirs, with the exception of jack pine needles, had rates of initial mass loss that were ~2 times greater compared to the same tissues placed in the unflooded forest sites (Figs. 3-5 to 3-9, 3-11 to 3-16). Mass loss in flooded jack pine needles was ~10% lower in the High C and Medium C reservoirs than in jack pine needles placed in corresponding unflooded forests. Mass loss in jack pine needles in the Low C reservoirs was similar to those placed in the Low C unflooded forest (Fig. 3-10).

Changes in C and N content

- Unflooded forests

By the end of the study, all plant tissues, with the exception of *Sphagnum* spp., placed in unflooded forest sites exhibited a gradual decline in %C, losing up to 20% of their original C over the three-year study (Table 3-3). Average %C content in *Sphagnum* spp. increased by 5% by the end of the study. %N increased by 5% to 100% in birch, blueberry, and Labrador tea leaves, bunchberry plants, jack pine needles, *Polytrichum* spp., *Pleurozium* spp, and lichen (Table 3-4) by the end of the three-year study. Average %N in alder leaves doubled after the first summer in all forest sites.

Table 3-3. Carbon content (%C) in plant tissue in litterbags sampled from unflooded forests and reservoirs. nd= no data, na= not applicable.

	High C		Medium C		Low C	
Tissue Date	Unflooded forest	Reservoir	Unflooded forest	Reservoir	Unflooded forest	Reservoir
Living trees						
Birch leaves						
Oct 99	51.6±0.4	55.3±0.2	51.4±0.7	55.5±0.1	53.9±0.2	56.3±0.6
May 00	51.2±0.4	54.7±0.8	53.0±0.1	52.4±1.7	52.0±0.5	55.6±0.4
Sept 00	49.3±0.03	52.4±0.4	52.3±0.4	52.1±0.2	50.7±0.2	52.3±0.8
May 01	43.0±5.8	50.5±1.1	47.9±1.3	49.1±2.3	42.0±9.3	49.5±0.8
Sept 01	41.2±4.4	48.4±0.8	49.4±0.5	47.5±0.5	50.6±0.3	48.3±2.5
Pine needles						
Oct 99	53.6±0.2	53.6±0.2	52.7±0.2	53.7±0.1	52.6±0.3	54.2±0.2
May 00	53.6±0.4	52.9±0.5	53.0±0.2	53.2±0.4	52.7±0.2	53.0±0.5
Sept 00	52.3±0.3	51.6±0.7	54.0±0.8	52.4±0.2	51.7±0.4	52.8±1.3
May 01	52.8±0.1	47.0±5.5	51.3±1.4	52.4±0.3	52.6±0.2	50.4±2.5
Sept 01	49.8±2.2	51.3±0.8	52.7±0.03	53.6±0.6	52.5±0.2	50.3±0.9
Wood						
Oct 99	50.9±0.6	50.4±0.6	50.6±0.4	50.3±0.1	50.5±0.1	51.6±0.6
May 00	50.00±0.6	50.6±0.3	52.0±1.7	50.4±0.2	51.9±0.4	50.8±0.2
Sept 00	49.0±0.3	49.6±1.0	50.1±0.1	49.1±1.2	49.1±0.7	49.1±0.7
May 01	47.8±2.8	49.2±0.3	49.6±0.6	48.4±1.5	51.5±1.4	49.4±0.6
Sept 01	47.8±0.9	48.8±0.8	49.9±1.0	47.0±2.0	48.6±1.2	50.4±0.4
Herbs and Shrubs						
Alder leaves						
Oct 99	52.9±0.2	52.0±2.2	54.7±2.1	55.4±0.3	54.1±0.5	55.2±0.1
May 00	51.5±0.7	55.3±0.3	52.5±0.04	54.2±0.7	52.3±1.4	54.8±0.3
Sept 00	48.0±1.6	52.6±0.4	52.6±0.1	51.6±1.0	51.7±0.1	51.9±0.6
May 01	35.6±15.2	51.0±0.4	48.4±1.3	50.1±1.5	42.6±4.7	49.1±1.2
Sept 01	40.3±9.1	50.6±1.6	49.0±2.1	50.1±1.2	51.8±0.2	49.0±0.8

Table 3-3. continued.

Tissue Date	High C Unflooded forest	High C Reservoir	Medium C Unflooded forest	Medium C Reservoir	Low C Unflooded forest	Low C Reservoir
Blueberry leaves						
Oct 99	53.1±1.1	54.1±0.3	52.8±0.2	55.5±0.4	54.8±0.4	54.5±0.4
May 00	54.6±0.1	54.2±0.4	52.9±0.6	52.6±0.8	53.6±0.4	54.2±0.1
Sept 00	52.2±0.3	50.0±1.5	53.3±0.6	52.7±0.6	53.5±1.9	52.0±0.4
May 01	50.9±1.6	50.8±0.3	51.6±0.5	51.1±0.9	52.4±1.3	49.0±0.8
Sept 01	49.8±2.1	50.2±0.5	52.5±0.3	48.5±1.6	52.7±0.5	48.6±0.7
Bunchberry plants						
Oct 99	48.3±0.1	49.4±2.4	46.6±0.1	52.0±0.3	nd	52.9±1.3
May 00	50.9±0.4	52.8±0.5	45.4±4.2	48.8±0.5	46.6±0.5	52.6±1.5
Sept 00	45.1±3.7	50.7±1.2	48.9±0.4	45.3±1.5	45.3±0.6	53.5±0.5
May 01	47.1±0.7	47.0±1.6	47.9±0.8	48.4±0.9	nd	38.1±2.8
Sept 01	46.6±1.9	52.7±0.4	41.4±7.2	43.6±2.4	47.9±0.4	51.1±0.01
Labrador tea leaves						
Oct 99	54.9±0.2	56.0±0.3	na	na	na	na
May 00	53.6±0.7	56.5±0.1	na	na	na	na
Sept 00	48.6±4.7	54.4±0.4	na	na	na	na
May 01	50.5±0.7	52.6±0.6	na	na	na	na
Sept 01	51.8±0.5	51.4±4.5	na	na	na	na
Bryophytes						
Sphagnum spp.						
Oct 99	46.2±0.5	44.3±0.2	na	na	na	na
May 00	41.0±0.2	45.2±0.2	na	na	na	na
Sept 00	43.3±0.4	40.8±0.4	na	na	na	na
May 01	43.5±0.2	41.9±0.4	na	na	na	na
Sept 01	48.9±6.3	40.5±0.5	na	na	na	na

Table 3-3. continued.

Tissue Date	High C Unflooded forest	High C Reservoir	Medium C Unflooded forest	Medium C Reservoir	Low C Unflooded forest	Low C Reservoir
Polytrichum						
Oct 99	na	na	na	na	47.6±0.4	48.9±0.4
May 00	na	na	na	na	49.1±0.5	48.4±0.2
Sept 00	na	na	na	na	46.4±0.1	44.3±1.6
May 01	na	na	na	na	46.8±0.3	45.8±0.4
Sept 01	na	na	na	na	46.4±0.1	42.5±1.2
Pleurozium						
Oct 99	na	na	na	na	48.1±0.2	47.8±0.2
May 00	na	na	na	na	47.9±0.5	47.4±0.2
Sept 00	na	na	na	na	46.6±0.1	44.9±0.3
May 01	na	na	na	na	47.1±0.5	44.6±0.2
Sept 01	na	na	na	na	44.5±1.6	43.8±0.3
Lichen						
Oct 99	na	na	na	na	43.5±2.8	46.6±1.6
May 00	na	na	na	na	45.5±6.1	47.0±1.6
Sept 00	na	na	na	na	43.8±0.1	43.3±0.2
May 01	na	na	na	na	24.7±11.2	39.9±2.1
Sept 01	na	na	na	na	39.2±5.2	41.6±0.4
'Old' wood						
Oct 99	na	na	59.6±0.1	59.3±0.1	na	na
May 00	na	na	59.8±0.1	58.5±0.6	na	na
Sept 00	na	na	59.8±0.2	57.2±0.4	na	na
May 01	na	na	58.8±0.2	56.5±0.5	na	na
Sept 01	na	na	57.3±0.9	57.3±0.5	na	na

Table 3-4. Nitrogen content (%N) in plant tissue in litterbags sampled from unflooded forests and reservoirs. nd= no data, na= not applicable.

	High C		Medium C		Low C	
Tissue Date	Unflooded forest	Reservoir	Unflooded forest	Reservoir	Unflooded forest	Reservoir
Living trees						
Birch leaves						
Oct 99	3.7±0.1	4.7±0.2	3.0±0.2	4.2±0.3	3.1±0.003	4.3±0.2
May 00	3.6±0.1	5.2±0.2	3.6±0.2	4.4±0.2	2.9±0.2	3.7±0.6
Sept 00	2.9±0.2	4.7±0.1	3.5±0.1	3.8±0.1	3.3±0.01	4.4±0.1
May 01	2.6±0.4	4.8±0.1	3.4±0.1	4.6±0.3	2.7±0.6	4.2±0.1
Sept 01	2.7±0.3	4.7±0.1	3.1±0.1	4.1±0.2	3.5±0.1	4.2±0.2
Pine needles						
Oct 99	1.4±0.2	1.5±0.04	1.4±0.04	1.5±0.02	1.3±0.01	1.5±0.04
May 00	1.8±0.03	1.5±0.1	1.5±0.02	1.6±0.1	1.4±0.6	1.4±0.04
Sept 00	2.0±0.1	1.7±0.1	2.0±0.3	1.7±0.1	1.5±0.1	1.8±0.1
May 01	2.3±0.1	1.7±0.2	2.0±0.1	1.8±0.1	1.7±0.2	1.8±0.1
Sept 01	2.5±0.2	1.7±0.04	2.1±0.03	2.1±0.1	1.9±0.02	1.8±0.1
Wood						
Oct 99	0.3±0.04	0.2±0.01	0.2±0.01	0.2±0.01	0.2±0.03	0.2±0.01
May 00	0.3±0.01	0.2±0.03	0.2±0.01	0.2±0.002	0.2±0.01	0.2±0.01
Sept 00	0.4±0.04	0.2±0.02	0.3±0.03	0.3±0.01	0.2±0.03	0.2±0.02
May 01	0.3±0.01	0.3±0.02	0.3±0	0.3±0.02	0.3±0.01	0.3±0.02
Sept 01	0.4±0.1	0.3±0.01	0.4±0.02	0.3±0.02	0.3±0.02	0.3±0.01
Herbs and Shrubs						
Alder leaves						
Oct 99	4.1±0.04	4.5±0.2	3.9±0.2	4.6±0.2	3.7±0.01	4.8±0.1
May 00	3.9±0.1	4.8±0.1	4.1±0.02	4.6±0.1	3.8±0.1	4.6±0.1
Sept 00	3.4±0.1	4.9±0.1	3.8±0.2	4.8±0.2	4.2±0.2	5.0±0.2
May 01	2.7±1.2	5.0±0.1	4.1±0.1	4.9±0.3	3.4±0.3	4.9±0.2
Sept 01	2.5±0.6	4.5±0.2	2.7±1.2	4.9±0.2	4.3±0.3	5.0±0.1

Table 3-4. continued.

Tissue Date	High C Unflooded forest	High C Reservoir	Medium C Unflooded forest	Medium C Reservoir	Low C Unflooded forest	Low C Reservoir
Blueberry leaves						
Oct 99	3.0±0.1	3.7±0.1	2.7±0.1	4.1±0.1	2.9±0.1	3.4±0.1
May 00	3.1±0.1	3.7±0.03	3.0±0.1	3.4±0.1	2.9±0.1	3.8±0.2
Sept 00	3.2±0.1	3.6±0.1	3.5±0.05	4.2±0.2	3.1±0.1	3.8±0.1
May 01	3.0±0.1	3.9±0.1	3.5±0.2	4.1±0.1	3.2±0.01	3.8±0.1
Sept 01	3.2±0.1	4.1±0.4	3.3±0.1	3.8±0.1	3.3±0.1	3.9±0.1
Bunchberry plants						
Oct 99	4.2±0.1	4.8±0.2	3.5±0.1	4.6±0.2	nd	4.3±0.3
May 00	3.7±0.1	5.1±0.2	3.8±0.4	3.7±0.1	3.5±0.1	3.1±0.3
Sept 00	3.4±0.2	4.1±0.2	3.9±0.1	3.3±0.3	0.3±0.3	3.5±0.1
May 01	3.7±0.1	4.2±0.2	3.3±0.1	3.9±0.3	nd	2.5±0.1
Sept 01	3.6±0.03	4.4±0.1	3.2±0.5	3.0±0.2	3.9±0.2	4.0±0.1
Labrador tea leaves						
Oct 99	1.9±0.03	2.2±0.04	na	na	na	na
May 00	2.1±0.1	2.2±0.1	na	na	na	na
Sept 00	2.1±0.2	2.3±0.1	na	na	na	na
May 01	2.3±0.02	2.6±0.1	na	na	na	na
Sept 01	2.3±0.1	2.3±0.1	na	na	na	na
Bryophytes						
Sphagnum spp.						
Oct 99	1.0±0.1	0.8±0.1	na	na	na	na
May 00	0.9±0.03	0.8±0.1	na	na	na	na
Sept 00	0.8±0.1	0.8±0.03	na	na	na	na
May 01	1.0	0.8±0.03	na	na	na	na
Sept 01	1.1±0.1	0.8±0.1	na	na	na	na

Table 3-4. continued.

Tissue Date	High C Unflooded forest	High C Reservoir	Medium C Unflooded forest	Medium C Reservoir	Low C Unflooded forest	Low C Reservoir
Polytrichum spp.						
Oct 99	na	na	na	na	1.7±0.02	1.3±0.1
May 00	na	na	na	na	1.7±0.02	1.2±0.1
Sept 00	na	na	na	na	1.6±0.1	1.1±0.1
May 01	na	na	na	na	1.7±0.1	1.2±0.03
Sept 01	na	na	na	na	1.6±0.1	1.1±0.1
Pleurozium spp.						
Oct 99	na	na	na	na	1.1±0.1	0.8±0.04
May 00	na	na	na	na	1.2±0.04	0.8±0.03
Sept 00	na	na	na	na	1.2±0.03	0.8±0.04
May 01	na	na	na	na	1.3±0.1	0.8±0.02
Sept 01	na	na	na	na	1.1±0.02	0.7±0.1
Lichen						
Oct 99	na	na	na	na	0.6±0.04	0.6±0.03
May 00	na	na	na	na	0.6±0.1	1.2±0.6
Sept 00	na	na	na	na	0.5±0.03	0.6±0.04
May 01	na	na	na	na	0.4±0.2	0.7±0.04
Sept 01	na	na	na	na	0.7±0.2	0.7±0.1
'Old' wood						
Oct 99	na	na	0.3±0.02	0.3±0.01	na	na
May 00	na	na	0.3±0.01	0.3±0.04	na	na
Sept 00	na	na	0.4±0.02	0.3±0.01	na	na
May 01	na	na	0.4±0.02	0.4±0.01	na	na
Sept 01	na	na	0.4±0.01	0.4±0.01	na	na

In the High C and Medium C forests, %N decreased to original values by the end of the study, whereas %N in tissues placed in the Low C forest continued to increase with time to values twice the original N content. There were no changes in the %N of old wood or *Sphagnum* spp. over time (Table 3-4). Changes in %C and %N of all plant tissues were similar among the three forested sites in all years.

- Reservoirs

Average %C in all plant tissues in reservoirs declined by up to 20% over the three-year study (Table 3-3). By the end of the study, %N increased up to 100% in flooded birch, alder, blueberry, and Labrador tea leaves, bunchberry plants, jack pine needles, wood, and lichen (Table 3-4). In flooded bryophytes, %N decreased by 10% in *Polytrichum* spp. and by 20% in *Sphagnum* spp. and *Pleurozium* spp. Changes in %C and %N of plant tissues placed in the three different reservoirs were similar and generally greater than in those in forests.

Changes in C and N mass

- Unflooded forests

Trends in C and N mass loss generally reflected changes in tissue mass. Birch, alder, blueberry, and Labrador tea leaves and bunchberry plants in unflooded forests exhibited gradual declines in C and N mass, with final losses as high as 80% and 70% of original C and N mass, respectively (Figs. 3-5 to 3-9). Bunchberry plants had the highest C and N mass loss, followed by birch and alder leaves (Fig. 3-9). Jack pine needles gradually lost C mass over time; however, N mass in jack pine needles remained at original values until the final year of study (Fig. 3-10). C and N mass in bryophytes and lichens also declined, but at slower rates than the more labile tissues (Figs. 3-11 to 3-14). Wood and old wood showed very little C loss, but exhibited up to 50 % increases in N (Figs. 3-15, 3-16).

- Reservoirs

In almost all cases, differences in C and N mass between tissues placed in deep and shallow sites in each reservoir were not statistically significant (Table 3-5), so we took the averaged the C and N mass in tissues placed in deep and shallow zones of each reservoir. Flooded birch, alder, blueberry, and Labrador tea leaves lost between 30% and 60% of their original C and 10% - 40% of their original N during the first summer of flooding (Figs. 3-5 to 3-9). Flooded bunchberry plants experienced the greatest loss of C and N mass (80 - 90% and 60 - 80% of original C and N lost, respectively) compared to all other tissues (Fig. 3-9). After an initial rapid decline in C mass in the first summer of flooding, C mass in submerged leaves and bunchberry plants stabilized, further declining only slightly by the end of the study.

Table 3-5. Results of ANOVA on carbon and nitrogen mass in litterbags taken from deep and shallow locations and from flooded and unflooded sites. Samples from all dates were tested together. * designates statistical differences at p=0.05.

Site	Deep versus Shallow sites		Flooded versus Unflooded sites	
	C mass	N mass	C mass	N mass
High C				
n	240	240	357	357
p value	0.539	0.081	0.808	0.008*
F value	0.38	3.09	0.21	4.94
Medium C				
n	208	208	312	312
p value	0.989	0.353	0.0001*	0.0001*
F value	0.001	0.87	43.56	29.14
Low C				
n	264	264	394	394
p value	0.080	0.011	0.0001*	0.0001*
F value	3.10	6.57	338.19	162.49

Carbon mass in flooded jack pine needles, bryophytes, and lichens consistently declined over the entire study period. Jack pine needles and bryophytes had initial C losses of 10% - 20% and final C losses ~30% of original

C mass (Figs. 3-10 to 3-13). Lichens lost ~50% of original C mass by the end of the study (Fig. 3-14). N mass in jack pine needles did not change over time (Fig. 3-10); bryophytes and lichens exhibited a 10% - 40% loss of N after the first summer of flooding (Figs. 3-5 to 3-8, 3-11 to 3-14). Wood and old wood exhibited very small percent of C loss over time (Figs. 3-15 to 3-16). N mass in wood and old wood increased over time to values ~10% and 35% greater than original N mass (Figs. 3-15, 3-16), respectively. Initial decomposition rates in flooded plants were greater than those in unflooded forests, the final C and N masses in tissues in forests and corresponding reservoirs were similar (see below).

Changes in C:N ratios

- Unflooded forests

Birch and alder leaves and bunchberry plants demonstrated sharp decreases of up to 40% in C:N ratios after the first summer (Figs. 3-5, 3-6, 3-9). C:N ratios in these tissues tended to stabilize after the first year and remained constant over time. C:N ratios in blueberry and Labrador tea leaves, jack pine needles, wood, and old wood had more constant declines over time and by the end of the study were generally 10 - 50% lower than original values (Figs. 3-7, 3-8, 3-10, 3-15, 3-16). C:N ratios in *Sphagnum* spp., *Pleurozium* spp., and lichens gradually increased to values ~20% higher than initial C:N ratios. Mid-way through the study C:N ratios in *Sphagnum* spp. and lichens decreased and were similar to pre-flood values by the conclusion of the study (Figs. 3-11, 3-14). C:N ratios in *Polytrichum* spp. decreased by less than 10% after the first season and were similar to initial values by the end of the study (Figs. 3-12, 3-13).

- Reservoirs

All flooded plant tissues, with the exception of bryophytes and lichens, had sharp 15% - 45% decreases in C:N ratios after the first summer of flooding (Figs.

3-5 to 3-10, 3-15, 3-16). C:N ratios in these tissues tended to stabilize after the first flooding season and remained constant over time with the exception of C:N ratios in flooded jack pine needles, wood, and old wood, which decreased more gradually over time. C:N ratios in flooded bryophytes (with the exception of *Pleurozium* spp.) and lichens increased by between 10% and 20% after the first summer of flooding. Whereas C:N ratios continued to increase in flooded *Sphagnum* spp. and *Polytrichum* spp. (Figs. 3-11, 3-13) after three summers, C:N ratios in flooded lichens gradually decreased by 20% over time (Fig. 3-13). C:N ratios in flooded *Pleurozium* spp. decreased by less than 10% after the first and second summer, but returned to initial values by the end of the study (Fig. 3-12).

Comparisons of C and N mass loss and C:N ratios in six tissues common to all sites

- Unflooded forests

Generally, for the six plant tissues placed in all three unflooded forests (birch, alder, and blueberry leaves, bunchberry plants, jack pine needles, and wood), those placed in the High C unflooded forest had the highest rates of C and N mass loss, followed by tissues in the Medium C forest. Tissues placed in the Low C forest had the least amount of C and N mass loss. The exception was wood, which did not exhibit differences in C loss among unflooded forest but did show a greater increase in N mass in the High C forest compared to the Medium C and Low C forests. C:N ratios did not differ in tissues placed among the three unflooded forests.

- Reservoirs

Carbon and N mass loss was greater in the High C reservoir compared to the Medium C and Low C reservoirs for four of the six tissues that were present in all three reservoirs (birch, alder, and blueberry leaves, and bunchberry plants; Figs. 3-5 to 3-7, 3-9). There was no apparent difference among reservoirs in loss of C and N from jack pine needles (Fig. 3-10) and wood blocks (Fig. 3-15). Inter-reservoir differences in C:N ratios were only apparent in bunchberry plants. C:N in bunchberry plants placed in the Low C reservoir were always higher than C:N in plants placed in the Medium C and High C reservoirs (Fig. 3-9).

Comparisons of C and N mass loss and C:N ratios in plants placed in unflooded forests and reservoirs

There were statistically significant differences in changes in C and N mass in tissues placed in the Medium C and Low C reservoirs compared to unflooded forests (p=0.0001 – 0.008, n=312 – 394). Changes in C mass in tissues in the High C forest were not significantly different from changes in C mass in tissues placed in the High C reservoir (p=0.81, n=357; Table 3-5). Initial rates of C and N mass loss from flooded birch, alder, blueberry, Labrador tea leaves, bunchberry plants, and jack pine needles in the Medium C and Low C reservoirs were greater than for those tissues placed in corresponding unflooded forest sites (Figs. 3-5 to 3-10). Although initial decomposition rates in flooded plants were greater than those in unflooded forests, the final C and N masses in tissues in forests and corresponding reservoirs were similar. C and N mass loss from *Sphagnum* spp., lichens, wood, and old wood placed in the reservoirs was similar to mass loss from these tissues in corresponding unflooded forests (Figs. 3-11 to 3-16). *Pleurozium* spp. and *Polytrichum* spp. mosses in unflooded forests exhibited an increase in C and N mass over time (Figs. 3-13, 3-14). As well, C:N ratios in these tissues increased over time at flooded sites, whereas C:N ratios in these tissues in did not change in the unflooded forest over time.

Changes in THg in decomposing litter in unflooded and flooded forests

THg concentrations

Concentrations of THg in plant tissues placed in both unflooded and flooded forests changed over time (Table 3-6). Generally, THg concentrations increased in plant tissues that had initial THg concentrations less than 30 ng g^{-1} (birch, alder, blueberry, and Labrador tea leaves, jack pine needles, and wood). In plant tissues with initial THg concentrations exceeding 30 ng g^{-1} (bryophytes and lichens), THg concentrations decreased over time (Table 3-6).

Changes in THg mass

- Unflooded forests

Overall, THg masses increased in birch, blueberry, alder, and Labrador tea leaves, jack pine needles, and wood over time. THg mass in birch and blueberry leaves in unflooded forest sites gradually increased to between 140% and 350% of original THg mass (Figs. 3-5, 3-6). After the first summer, THg masses in alder leaves, jack pine needles, and wood decreased by up to 40%. In subsequent seasons, THg mass in jack pine needles and wood in all forest sites increased to final values that were 113% - 154% and 150% - 281% higher than original THg masses, respectively; (Figs. 3-10, 3-15). By the end of the study, THg mass in alder leaves in the Medium C and Low C forests increased to 149% and 139% of that originally found in the tissues, respectively, whereas THg mass in alder leaves in the High C forest decreased to 81% of their original THg mass. THg mass in Labrador tea leaves remained relatively constant after the first summer (103% of original THg mass), decreased after the second summer (68% of original THg mass), and increased after the third summer (117% of original THg mass; Fig. 3-8). THg mass in old wood and bunchberry plants did not change over time, with the exception of THg mass in bunchberry plants placed in the High C forest, which decreased ~20% from original THg mass (Fig. 3-9). We underestimated the amount of bunchberry plants required and therefore bunchberry plants were not sampled from the High C forest in the third summer.

Table 3-6. Total mercury concentrations (ng THg g^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Tissue Date	High C Unflooded forest	 Reservoir	Medium C Unflooded forest	 Reservoir	Low C Unflooded forest	 Reservoir
Living trees						
Birch leaves						
Oct 99	17.51	47.99±20.01	11.60	33.10±7.67	10.69	36.40±1.91
Sept 00	34.70	54.51±15.71	42.16	50.52±14.51	15.26	44.33±0.42
Sept 01	47.53	64.26±4.06	59.08	82.23±30.66	16.12	54.64±13.8
Pine needles						
Oct 99	15.92	15.30±2.40	13.75	14.30±0.67	18.61	15.99±1.25
Sept 00	26.61	18.21±1.04	22.38	19.13±2.14	16.78	17.37±2.21
Sept 01	43.03	26.51±1.23	39.54	26.49±2.41	23.65	23.53±1.41
Wood blocks						
Oct 99	3.27	3.47±0.67	2.67	4.18±0.28	3.10	2.82±0.38
Sept 00	12.21	6.41±0.16	4.61	5.56±1.06	5.34	5.99±1.34
Sept 01	11.72	5.59±0.02	7.71	4.58±0.07	5.82	4.01±0.27
Herbs and Shrubs						
Alder leaves						
Oct 99	14.56	35.78±5.27	14.47	29.21±1.79	10.06	26.99±0.68
Sept 00	33.34	43.67±8.54	33.86	40.98±0.45	14.93	40.84±1.19
Sept 01	30.43	52.85±3.70	41.00	52.71±10.82	30.50	50.12±0.81
Blueberry leaves						
Oct 99	12.50	35.89±12.15	11.17	24.05±0.71	9.19	27.85±0.67
Sept 00	24.13	43.45±4.16	26.71	47.25±9.18	21.71	35.21±2.64
Sept 01	33.02	57.53±10.15	40.40	53.59±12.65	26.46	46.59±9.12
Bunchberry plants						
Oct 99	16.18	88.61±29.37	14.95	77.88±8.65	nd	89.56
Sept 00	37.71	60.99±11.50	42.14	70.63±5.79	27.92	72.92
Sept 01	48.07	62.49±10.04	53.03	102.34±36.92	46.67	73.48

Table 3-6. THg concentrations (ng g^{-1}) continued.

	High C		Medium C		Low C	
Tissue Date	Unflooded forest	Reservoir	Unflooded forest	Reservoir	Unflooded forest	Reservoir
Labrador tea leaves						
Oct 99	33.87	41.06±8.02	na	na	na	na
Sept 00	30.21	41.80±0.34	na	na	na	na
Sept 01	66.36	57.25±5.69	na	na	na	na
Bryophytes						
Sphagnum spp.						
Oct 99	56.41	32.94±10.38	na	na	na	na
Sept 00	38.49	30.34±3.20	na	na	na	na
Sept 01	47.35	28.22±3.83	na	na	na	na
Polytrichum spp.						
Oct 99	na	na	na	na	50.16	42.53±3.01
Sept 00	na	na	na	na	60.86	38.63±4.99
Sept 01	na	na	na	na	74.77	44.71±5.27
Pleurozium spp.						
Oct 99	na	na	na	na	58.72	38.44±2.37
Sept 00	na	na	na	na	66.38	39.56±5.93
Sept 01	na	na	na	na	54.00	45.30±10.19
Lichen						
Oct 99	na	na	na	na	26.08	29.78±0.45
Sept 00	na	na	na	na	29.49	22.64±0.03
Sept 01	na	na	na	na	24.91	23.95±6.34
'Old' wood						
Oct 99	na	na	7.01	9.30±0.99	na	na
Sept 00	na	na	7.94	12.54±2.48	na	na
Sept 01	na	na	9.35	15.26±0.42	na	na

The mass of THg in bryophytes and lichen placed in forests decreased over time. With the exception of *Sphagnum* spp., THg mass in these tissues decreased between 10% and 50% of original THg after the first summer and remained relatively constant in subsequent seasons. *Sphagnum* spp. lost 6% of their original THg mass after the first summer and an additional 36% in the second and third summers (Fig. 3-12).

- In reservoirs

As with C and N mass, THg mass in plants in deep and shallow zones were averaged to obtain THg masses that were representative of each reservoir. THg mass increased in flooded birch, alder, blueberry, and Labrador tea leaves, jack pine needles, wood, and old wood. The greatest increases in THg mass were observed in submerged birch and blueberry leaves. Average THg mass in these tissues increased sharply after the first summer of flooding and continued to increase during each flooding summer (Figs. 3-5, 3-7). Average increases in THg mass were between 61.3 - 107 ng for birch leaves and 52.7 - 124.6 ng for blueberry leaves, corresponding to levels between 167% and 428% of original THg mass. The mass of THg also increased in flooded alder and Labrador tea leaves, jack pine needles, wood, and old wood; however, THg increases in these tissues tended to occur more gradually over time compared to birch and blueberry leaves. Final increases in THg mass in alder leaves (124% - 172% of original; Fig. 3-6), wood (103% - 149% of original; Fig. 3-15), and old wood (151% of original; Fig. 3-16) were greater than in Labrador tea leaves and jack pine needles (116% and 113% - 125% of original, respectively; Figs. 3-8, 3-10). There was an initial increase in THg mass in bunchberry plants after the first flooding summer, followed by gradual declines in THg mass, with final average levels between 57% and 80% of original THg mass (Fig. 3-9).

We observed decreases in THg mass in bryophytes and lichen placed in the upland reservoirs. THg loss from bryophytes was greatest during the first summer of flooding and remained relatively constant over time. *Sphagnum* spp., *Pleurozium* spp., and *Polytrichum* spp. lost between 41% and 64% of their original THg mass in the first summer, but lost very little THg mass thereafter. Lichens lost 32% of their original THg mass after the first summer of flooding, and then gradually lost another 36% of their THg mass by the end of the study.

Comparisons of THg mass in six tissues common to all sites

- Unflooded forests

In birch, alder, and blueberry leaves, and jack pine needles, final increases in THg mass were between 1.1 and 2.2 times greater in the Medium C forest compared to the High C and Low C forests (Figs. 3-5 to 3-7, 3-10). In wood, the largest increase in THg mass was observed in the High C forest (Fig. 3-15). There were no differences in THg mass in bunchberry plants placed among the three unflooded forests (Fig. 3-9).

- Reservoirs

The greatest increases in THg mass in flooded birch, blueberry, and alder leaves were observed in the High C reservoir, generally followed by increases in THg mass in these tissues in the Medium C, and then Low C, reservoirs. Final THg mass in submerged birch, blueberry, and alder leaves in the High C reservoir were between 1.3 and 1.7 times higher than THg mass in the same tissues placed in the Medium C and Low C reservoirs, respectively. In tissues that did not exhibit an increase in THg mass (bunchberry plants, jack pine needles, and wood), there were no differences in THg mass among plants placed in the three reservoirs.

Comparisons of THg mass in plants in unflooded forests and reservoirs

Generally, initial rates of change in THg mass over time were greater in tissues in reservoirs than in the same tissues in forests (Figs. 3-7, 3-10).

However, there were few differences in final THg mass in the birch, blueberry, alder, and Labrador tea leaves, bunchberry plants, and jack pine needles placed in reservoirs compared to those placed in forests (Figs. 3-5 to 3-9). The THg mass in old wood increased more in the Medium C reservoir than in the forest adjacent to the reservoir (Fig 3-16). Final THg mass in bryophytes and lichens in reservoirs were between 1.2 and 2.4 times less than final THg mass in the same tissues in their corresponding forests (Figs. 3-11 to 3-14). There were no differences in THg mass in wood in reservoirs compared to wood in forests, with the exception of wood in the High C forest, which had higher THg mass compared to wood in the High C reservoir (Fig. 3-15).

Changes in MeHg in decomposing litter in unflooded and flooded forests

MeHg concentrations

MeHg concentrations in blueberry and Labrador tea leaves, jack pine needles, *Polytrichum* spp., and lichen placed in unflooded forests decreased after the first summer to concentrations that were up to 60% lower than initial concentrations (Table 3-7). By the end of the study, MeHg concentrations in jack pine needles and blueberry leaves increased to values that were up to 11 times higher than initial MeHg concentrations. MeHg concentrations in Labrador tea leaves, *Polytrichum* spp., and lichen increased at the end of the study, but final MeHg concentrations did not exceed initial concentrations (Table 3-7). MeHg concentrations in *Pleurozium* spp. increased by ~45% after the first summer, but by the end of the study had decreased to concentrations that were almost half initial concentrations.

MeHg concentrations in flooded jack pine needles, blueberry and Labrador tea leaves, *Polytrichum* spp., *Pleurozium* spp., and lichen increased the first summer of flooding to values that were between 2 and 28 times higher than initial concentrations. MeHg concentrations remained elevated over initial MeHg concentrations until the end of the study (Table 3-7). The greatest increases in MeHg concentrations were observed in blueberry and Labrador tea leaves, which increased up to 36 times initial concentrations. *Pleurozium* spp. and lichens displayed the smallest increases in MeHg concentrations, increasing only 3 times initial values.

Table 3-7. Methylmercury concentrations (ng MeHg g^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs.

	High C		Medium C		Low C	
Tissue Date	Unflooded forest	Reservoir	Unflooded forest	Reservoir	Unflooded forest	Reservoir
Living trees						
Pine needles						
Oct 99	0.081	0.724	0.040	0.272	0.077	0.358
Sept 00	below detection	0.676	0.130	0.069	0.071	0.519
Sept 01	0.936	1.104	0.274	1.656	0.280	0.765
Herbs and shrubs						
Blueberry leaves						
Oct 99	0.847	3.645	0.135	1.519	0.113	3.402
Sept 00	0.163	4.376	0.227	3.913	0.231	2.173
Sept 01	0.701	0.841	0.229	6.558	0.255	5.303
Labrador tea leaves						
Oct 99	0.430	1.371	na	na	na	na
Sept 00	0.543	1.950	na	na	na	na
Sept 01	0.281	6.216	na	na	na	na
Bryophytes						
Polytrichum spp.						
Oct 99	na	na	na	na	0.144	1.965
Sept 00	na	na	na	na	0.469	4.045
Sept 01	na	na	na	na	0.396	5.690
Pleurozium spp.						
Oct 99	na	na	na	na	1.879	5.614
Sept 00	na	na	na	na	0.604	2.328
Sept 01	na	na	na	na	0.917	4.568
Lichen						
Oct 99	na	na	na	na	0.362	0.993
Sept 00	na	na	na	na	0.098	0.769
Sept 01	na	na	na	na	0.363	1.565

MeHg mass

- In unflooded forests

After the first and second summers of study, MeHg mass generally decreased in blueberry and Labrador tea leaves, jack pine needles, *Polytrichum* spp., and lichen (Figs. 3-7, 3-8, 3-10, 3-13, 3-14). By the end of the study, MeHg mass in these tissues increased, but only jack pine needles had MeHg mass increases that exceeded initial values. Final MeHg masses in jack pine needles were 63% and 294% higher than initial MeHg mass (Fig. 3-10). Increases in MeHg mass in *Pleurozium* spp. were observed after the first summer, but by the end of the study, feather mosses had lost 46% of their initial MeHg mass (Fig. 3-12).

- In reservoirs

The largest increases in MeHg mass were observed in flooded blueberry leaves, where MeHg mass increased by between 322% and 1378% after the first summer of flooding (Fig. 3-7). Flooded jack pine needles also exhibited large increases in MeHg mass over time. After the first summer of flooding, MeHg mass in jack pine needles was between 150% and 572% higher than initial masses (Fig. 3-10). MeHg mass in Labrador tea leaves, *Pleurozium* spp., and *Polytrichum* spp. increased by 96%, 230%, and 285%, respectively, by the end of the first season of flooding (Figs. 3-8, 3-12, 3-13). The increase in MeHg mass was slowest in flooded lichen during the first flooding season (only 34% higher than the original MeHg mass; Fig. 3-14). After the first season of flooding, MeHg mass in blueberry and Labrador tea leaves, jack pine needles, and *Polytrichum* spp. continued to increase with time to final values as high as 2030% of original MeHg masses (Figs. 3-7, 3-8, 3-10, 3-13). Final MeHg mass in flooded *Pleurozium* spp. and lichens decreased to 250% and 89% of original MeHg mass (Fig. 3-12, 3-14).

Comparisons of MeHg mass in tissues common to all sites

After the first summer, decreases in MeHg mass in blueberry leaves and jack pine needles were similar in all three forests, with the exception of blueberry leaves in the High C forest, which exhibited a 320% increase in MeHg mass (Figs. 3-7, 3-10). At the end of the study, MeHg mass in blueberry leaves increased slightly in the Medium C and Low C forests, but did not exceed original MeHg masses. At the end of the study, blueberry leaves in the High C forest were ~180% higher than original MeHg mass (Fig. 3-7). Final MeHg mass in jack pine needles increased in all three forests, and was highest in the High C forest compared to the Medium C and Low C forests (Fig. 3-10).

After the first summer of flooding, increases in MeHg mass in blueberry leaves and jack pine needles were highest in the High C reservoir (1360% and 710% of original MeHg mass in blueberry leaves and jack pine needles, respectively), and lowest in the Medium C reservoir (330% and 260% of original MeHg mass in blueberry leaves and jack pine needles, respectively; Figs. 3-7, 3-10). By the end of the study, however, increases in MeHg mass in flooded jack pine needles and blueberry leaves the Medium C sites exceed those in the other reservoirs.

Comparisons of MeHg mass in plants in unflooded forests and reservoirs

Rates of increase in MeHg mass in blueberry and Labrador tea leaves, jack pine needles, *Pleurozium* spp., *Polytrichum* spp, and lichen were higher in the reservoirs compared to unflooded forests. Lichen were the only plant tissue that lost MeHg over time. MeHg mass in blueberry and Labrador tea leaves, jack pine needles, *Pleurozium* spp., and *Polytrichum* spp. increased substantially, whereas all plant tissues tested, with the exception of jack pine needles, in unflooded forests lost MeHg.

Discussion

Decomposition of litter in unflooded and flooded forests

Plant tissues in unflooded forests and reservoirs decomposed as indicated by changes in tissues mass, C and N mass, and C:N ratios. In all of the sites, large differences in decomposition rates among our 12 plant tissues were observed. Generally, decomposition rates followed the order: herbs (bunchberry) > deciduous leaves (birch, alder, blueberry, Labrador tea) > jack pine needles > bryophytes and lichens > wood and old wood. For like tissues, we also observed faster initial decay rates in reservoirs compared to unflooded forest sites; however, C and N losses after ~800 days were similar in unflooded and flooded plant tissues.

Decomposition of plants in unflooded forests

Tissue decomposition rates in unflooded forest sites were similar to those found in other studies examining plant tissue decomposition in the boreal forest ecoregion (Meentemeyer 1978; Melillo and others 1982; Moore 1984; Taylor and others 1989; Moore and others 1999; Prescott and others 2000; Thomas and Prescott 2000; Spänhoff and others 2001; Thormann and others 2001; Morrison 2003). Two main factors affect leaf litter decomposition in terrestrial ecosystems (Swift and others 1979). The first is the micro- and macro- climates (most notably temperature and moisture conditions; Aerts 1997) and light; Caldwell and others 1998; Zepp and others 1998) to which the litter is exposed. The second factor is related to the chemical nature of the litter, or litter quality. These two factors interact to determine decomposition rates (Swift and others 1979; Melillo and others 1989). Globally, the effects of litter chemistry on plant decomposition are secondary to the effects of climatic conditions (Aerts 1997). Locally, however, where decomposing plants experience similar climates, litter quality primarily determines litter decomposition.

Litter quality is often defined by the C:N ratio in plant tissues (Swift and others 1979). Terrestrial autotrophs generally have low N content and C:N ratios in terrestrial plants range from 7.5 and 225, with a mean of 36 (Elser and others 2000; Sterner and Elser 2002). Initial C:N ratios in our plant tissues are within published ranges, with a low of 18.4 for alder leaves, a shrub capable of N_2 fixation, and a high of 226 for wood. Studies have found tissues with large C:N ratios (greater than 25) experience little net mineralization of N and therefore decompose slowly over time (Meentemeyer 1978; Swift and others 1979; Webster and Benfield 1986; Hirschel and others 1997). Based on initial C:N ratios in our plant tissues, we expected to see higher decomposition rates in the herb, birch, alder, and blueberry leaves, very little decomposition in wood and old wood, and intermediate rates in the other tissues. Our data supported this hypothesis.

Over time, C:N ratios decreased in the herb, deciduous leaves, jack pine needles, wood, and old wood; increased in *Sphagnum* and lichens; and did not change in *Polytrichum* spp. and *Pleurozium* spp. mosses. Generally, C:N ratios in our plant tissues decreased over time if they had an initial N content of between 1.3 – 2.8 %N and increased if initial N content was lower (0.1 - 1.5 %N; Table 3-4). The exception was wood and old wood, which showed decreases in C:N ratios, but had extremely low N content (0.2 %N and 0.03 %N, respectively) compared to other plant tissues. Our results were expected because the more N a plant contains, the more rapidly it will decay (Melillo and others 1984).

Plants that increase in C:N ratios lose N more quickly than C and therefore represent a valuable source of N to the forest floor. Plants that decrease in C:N ratios lose C more quickly than N, which suggests either loss of C or significant amounts of N immobilization (Knops and others 2002). N immobilization is defined as the increase in the absolute amount of N in plant tissues through the accrual of N into microbial biomass, as well as the accumulation of N in products of microbial activity (Melillo and others 1984). This requires the addition of N to decomposing tissues from surrounding environments (Melillo and others 1984). In our study, both C and N mass decreased in decomposing birch, alder, blueberry and Labrador tea leaves, bunchberry plants, jack pine needles, bryophytes, and lichen. These tissues contributed C and N to the surrounding forest, whereas N mass increased in wood and old wood and removed N from the forest sites.

The effects of environmental conditions on decomposition were apparent when we examined differences in C and N mass loss in tissues placed in the High C unflooded forest compared to those placed in the Medium C and Low C unflooded forest sites. C and N mass loss from tissues placed in the High C forest were greater than decay rates of the same tissue type placed in the other forest sites. This is likely due to the placement of litterbags in a hollow in the High C forest, which often had higher moisture conditions than the other sites. The wet saturated environment in the hollow in the High C forest likely promoted increased rates of decomposition, because decay occurs most rapidly in environments where biotic activity is highest (Schlesinger 1997). As well, water facilitates the movement of decomposition by-products from plant tissues to surrounding environments. It also possible that the environment in this saturated hollow was anaerobic and resulted in faster decay rates of plant tissues compared to those decomposing in an aerobic environment (Melillo and others 1984). The Medium C and Low C unflooded forest sites were well drained and there were no differences between the rates of decomposition in plants placed in these two sites.

Decomposition of plant tissues in reservoirs

Leaf breakdown occurs more rapidly in freshwater systems than in terrestrial systems (Webster and others 1986) and therefore we expected plant tissues placed in reservoirs to decompose more rapidly than tissues placed in the unflooded forests. We observed faster rates of plant tissue mass loss, and C and N mass loss, in flooded tissues compared to those in unflooded forests in the early period of the experiment. There are three distinct phases in the breakdown of vascular plants in water. The first is rapid, initial loss of soluble and labile organic material through leaching. This is followed by a period of microbially mediated decomposition, which involves the immobilization of elements (especially N and P) into bacteria and bacterial by-products. The final stage is mechanical and invertebrate fragmentation (Webster and others 1986). Once organic matter is degraded to the small chain fulvic and humic acids that define dissolved organic carbon, photochemical degradation becomes important in mineralizing carbon (Schiff and others 1997; Häder and others 1998). Elevated decomposition rates in plants in reservoirs occurred in the first 79 days after flooding. After the first summer of flooding, decomposition rates as measured by tissue mass loss and C and N mass loss were no different in submerged plants compared to unflooded plants. High decay in the first summer of flooding indicated increased leaching of C and N from submerged plant tissues compared to unflooded plant tissues. Microbial mediated decomposition occurs over a longer time period; therefore, little difference between decomposition rates in submerged and unflooded plant tissue was expected over the long term.

Decreases in C:N ratios and increases in N mass indicate that jack pine needles, *Pleurozium* spp., wood, and old wood were removing N from the reservoirs during N immobilization. Submerged birch, blueberry, alder, and Labrador tea leaves, bunchberry plants, *Sphagnum* spp., *Polytrichum* spp., and lichens lost both C and N mass, suggesting that these tissues were sources of C and N to the reservoirs.

In aquatic systems, litter quality is considered the primary controller of decomposition rates. However, environmental factors such as temperature and dissolved nutrient concentrations are also important (Melillo and others 1984; Webster and others 1986). Generally, litter decomposition is accelerated in warmer waters that have higher concentrations of dissolved N and P (Melillo and others 1984; Royer and Minshall 2001). There were no differences in changes in C and N mass among reservoirs, which suggests that environmental factors affecting decomposition did not differ among the three upland reservoirs. In fact, there were little differences in water column (Chapter 4, this thesis) and soil (Matthews and others submitted) temperatures and dissolved N and P concentrations (R.A. Bodaly and A. Majewski, unpublished data) among reservoirs within a given year.

Total Mercury

THg mass in birch, blueberry, alder, and Labrador tea leaves, jack pine needles, and wood in both unflooded and flooded forests and in old wood in reservoirs increased with time. Rates of THg mass increases were greatest in birch and blueberry leaves, whereas THg mass in bunchberry plants remained relatively constant with time. This is the first study examining THg dynamics in decomposing upland plant tissues placed in unflooded and flooded forests and the first to show increases of THg in decomposing plant tissue.

THg mass in bryophytes and lichens decreased in unflooded and flooded plant tissues. Decreases in THg in *Sphagnum* spp. were supported by results from a previous study by Heyes and others (1998) that found THg mass decreased in *Sphagnum fuscum* placed in litterbags in pristine and impounded wetlands. However, they also observed decreases in THg mass in other plant tissues, *Carex rostrata* (grass) stems and *Picea mariana* (spruce) needles.

What factor controls changes in THg mass in decomposing plant tissues?

In both unflooded and flooded sites, THg mass increased in plant tissues with initial THg concentrations that were less than 30 ng g^{-1} and decreased in plant tissues with initial THg concentrations exceeding 30 ng g^{-1}, suggesting that changes in THg mass depend on initial THg concentrations. It was expected THg mass in jack pine needles in our study to decrease like the THg mass in spruce needles reported in Heyes and others (1998), but here THg mass increased in jack pine needles. However, initial THg concentrations in jack pine needles (14.1 ng g^{-1}) were approximately half the initial THg concentrations in spruce needles (27.5 ng g^{-1}). We conclude that THg concentrations in decomposing plant tissues in natural forests and reservoirs equilibrate with THg concentrations in surrounding environments, and that initial THg concentrations control changes in THg mass in decomposing tissues.

We did not sample soil concentrations in the forest sites, but concentrations of THg in organic soils in the High C, Medium C, and Low C reservoirs were measured prior to flooding from cores collected using a stainless steel barrel corer lined with plastic sleeves (see Chapter 4; this thesis). THg concentrations were much higher in organic soils (89, 44, and 39 ng cm^{-2} d.w.; unpublished data, K.R. Rolfhus, University of Wisconsin at La Crosse, La Crosse, WI) than in decomposing plants. If THg in plants equalized with THg in soils only, we would expect to see greater increases in plants in the High C forest, compared to those in other sites. However, processes occurring in unflooded forest sites were dynamic and precipitation, throughfall, and runoff, in addition to organic soils, could have been sources of THg to decomposing plants.

Differences in changes in plant tissue THg concentrations among reservoirs may be due to differences in THg concentrations in the water and flooded soils surrounding the litterbags. THg concentrations in water at the flooded soil-water interface were higher in the High C reservoir (13.3, 2.6, and 2.5 ng L^{-1} in September in years 1 - 3), than in the Medium C (3.2, 1.9, and 1.7 ng L^{-1} in September in years 1 - 3) and Low C (3.3, 2.2, and 1.1 ng L^{-1} in September in years 1 - 3) reservoirs (unpublished data, K.R. Rolfhus; Appendix 16). Although we have no data on THg concentrations in flooded soils, we assumed based on pre-flood values, that THg concentrations in the organic soils in the High C reservoir were greater than those in the Medium C and Low C reservoirs. We conclude that THg concentrations in decomposing plant tissues in the aquatic systems equilibrated with flooded soils and overlying water concentrations.

Differences in changes in THg mass in unflooded forests and reservoirs

For plant tissues decomposing in both forests and reservoirs, changes in THg mass were dictated by the combined effect of tissue mass loss and equilibrium with concentrations of THg in surrounding soils and water. However, although there were no differences in C and N loss among the three different forests and reservoirs, changes in THg mass did differ among the High C, Medium C, and Low C sites.

Generally, rates of change in THg mass were greater in flooded plants than in the same species of unflooded plants. These results are consistent with those of Heyes and others (1998), which found greater THg decreases in tissues in impounded wetlands compared to tissues in non-impounded wetlands. However, for plant tissues that increased in THg mass, there were few differences in final THg masses in flooded plants compared to unflooded plants. This suggests that THg mass in flooded plants reached equilibrium with reservoir water faster than unflooded plants did with the surrounding forest floor, but that equilibrium was reached in both unflooded and flooded sites within the 800-day study period. For plants that showed a decrease in THg mass (bryophytes and lichens), final THg values in submerged plants were less than plants in the corresponding unflooded forest. This suggests that bryophytes and lichens in the unflooded forests had not equilibrated with the surrounding forest by the end of the study.

Methylmercury

Initial MeHg concentrations were similar to those found in an earlier survey of MeHg and THg in plants at the ELA (Moore and others 1995). General trends indicate that, over time, MeHg mass in blueberry and Labrador tea leaves, jack pine needles, *Pleurozium* spp., and *Polytrichum* spp. decreased in unflooded forests (with the exception of the High C forest) and increased in reservoirs. These results are consistent with those reported by Heyes and others (1998) who found that MeHg mass increased in plant tissues (spruce needles, sedge grasses, and *Sphagnum* spp.) placed in impounded wetlands compared to dry wetland sites.

Decreases in MeHg mass in plants in the Medium C and Low C forests indicate a loss of MeHg from plant tissues decomposing in unsaturated, well-drained forest soils. Decomposing plants therefore possibly represented an input of MeHg to soils in unflooded forests, which would then presumably be available for transport to aquatic systems. MeHg concentrations in unflooded soils were 0.20 and 0.52 ng g-2 (unpublished data, K.R. Rolfhus) in the Medium C and Low C forests, respectively, which were similar to MeHg concentrations in decomposing plant tissues. It is also possible that since MeHg production is suppressed and MeHg degradation enhanced in aerobic environments (Ullrich and others 2001), MeHg in plant tissues in unflooded forests was destroyed by microbial demethylation.

We observed increases in MeHg in plant tissues placed in the High C forest (the wet saturated hollow) and in flooded forests. A number of studies have shown that inundation of plant tissue results in increased MeHg concentrations in surrounding water and biota (Chapter 2, this thesis; Hecky and others 1991; Porvari and Verta 1995; Kelly and others 1997; Heyes and others 1998; Thérien and Morrison 1999; Balogh and others 2002; Chapter 4, this thesis; St. Louis and others submitted). It is also possible, although unlikely, that increases in MeHg mass in plant tissues placed in saturated or flooded environments, were due to uptake of MeHg from the surrounding water and soils. However, increases in MeHg mass more likely reflect increased rates of methylation in and on plant tissues. Whole ecosystem MeHg input-out budgets in reservoirs (presented in Chapter 4, this thesis) and a sealed enclosure experiment examining methylation in jack pine needles and birch leaves (Chapter 2, this thesis) clearly showed that increases in MeHg were due to increased methylation rates.

At present, we do not have sufficient data to determine if rates of increases in MeHg in flooded plant tissues were consistently higher in one reservoir compared to the others. However, differences in MeHg increases in plant tissues may explain the differences we observed in MeHg concentrations in reservoir water (see Chapter 4, this thesis). In our reservoirs, much of the leaves and needles below the water line remained attached to the plants (personal observation) and were therefore situated in the water column. The position of flooded plant tissue in the water column possibly facilitated fluxes of MeHg from areas of active methylation (plants) to the water column. As well, plants decomposing in reservoirs could be an important source of MeHg to aquatic organisms. Because the majority of MeHg in fish comes from their diet (Hall and others 1997), aquatic invertebrate grazing on flooded, decomposing plant tissues and the bacteria associated with them could be an effective vector for the transfer of MeHg to higher trophic organisms.

Conclusions

While initial rates of decomposition were slower in tissues placed in unflooded forests compared to the same tissues placed in reservoirs, there were no differences in the amount of C and N mass left in the tissues at the end of the study. THg mass increased in flooded birch, alder, blueberry, and Labrador tea leaves, jack pine needles, wood, and old wood and there were few differences between plants in forests compared to reservoirs (Table 3-8). In tissues in which THg declined, the rate of loss was higher in flooded plants compared to the same plant tissue placed in unflooded forests, but there were no differences in final THg masses in unflooded forests and reservoirs. Changes in THg mass resulted from the combined effects

Table 3-8. Approximate ranges of total mercury (THg) and methylmercury (MeHg) mass remaining (percent of original mass) in plant tissues from litterbags sampled from unflooded forests and reservoirs.

	Unflooded forest		Reservoir	
Tissue	%THg mass remaining	%MeHg mass remaining	%THg mass remaining	%MeHg mass remaining
Living trees				
Birch leaves	120 – 300%	nd	230 – 380%	nd
Pine needles	90 – 150%	40 – 400%	80 – 130%	260 – 1200%
Wood blocks	70 – 300%	nd	80 – 170%	nd
Herbs and Shrubs				
Alder leaves	60 – 140%	nd	80 – 200%	nd
Blueberry leaves	120 – 350%	50 – 120%	160 – 500%	300 – 1300%
Bunchberry plants	70 – 300%	nd	60 – 300%	nd
Labrador tea leaves	70 – 120%	30 – 80%	80 – 130%	200 – 800%
Bryophytes				
Sphagnum spp.	60 – 90%	nd	40 – 80%	nd
Polytrichum spp.	50 – 60%	30 – 100%	30 – 40%	400 – 1000%
Pleurozium spp.	90%	40 – 150%	50%	140 – 400%
Lichen	80%	20– 70%	30 – 70%	80 – 130%
‘Old’ wood	75 – 100%	nd	100 – 150%	nd

of decomposition and the establishment of an equilibrium between THg concentrations in decomposing plant tissues and THg concentrations in surrounding soils and water. Bryophytes and lichens were sources of THg to unflooded and flooded forests, while birch, blueberry, alder, and Labrador tea leaves, jack pine needles, and wood took up THg from surrounding environments.

Plant tissues placed in unflooded forests lost MeHg to their surrounding environments and were possibly sources of MeHg to the forest floor, although MeHg could have subsequently been demethylated to inorganic Hg. Although there were few differences in decomposition and THg mass increases in plants placed in unflooded forests compared to flooded forests, we observed substantive increases in MeHg mass in reservoirs compared to unflooded forests, likely due to increased rates of net MeHg production. Flooded plant tissue may be an important source of MeHg to grazing invertebrates and a probable avenue of transfer of MeHg to fish.

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Chapter 4. Impacts of reservoir creation on the biogeochemical cycling of methyl and total mercury in boreal upland forests

Introduction

An important environmental consequence of flooding landscapes and creating reservoirs is the bioaccumulation of methylmercury (CH_3Hg^+; MeHg), a strong vertebrate neurotoxin, through the food web into fish. The health of people depending on reservoir fisheries for food is of concern, because human consumption of fish with sufficiently high concentrations of MeHg may cause teratogenic effects and irreversible neurological damage (Kurland and others 1960; Myers and others 2000; Rosenberg and others 1997). For example, MeHg concentrations in predatory fish harvested from northern Boreal reservoirs in Manitoba (Bodaly and others 1984; Hecky and others 1991), Québec (Brouard and others 1994; Schetagne and Verdon 1999), and Newfoundland (Scruton and others 1994), Canada and Finland (Lodenius and others 1983) often exceed Canadian marketing limits of 0.5 $\mu g\ g^{-1}$ wet mass for more than 20 years after initial flooding. It is therefore imperative that stakeholders consider MeHg contamination of fisheries when planning the construction of new reservoirs.

The decomposition of organic carbon (OC) in flooded vegetation and soils in reservoirs fuels the microbial methylation of inorganic mercury (HgII; IHg) to MeHg (Compeau and Bartha 1984; Hecky and others 1991; Kelly and others 1997; Chapter 2, this thesis), as well as the production of the greenhouse gases (GHGs) carbon dioxide (CO_2) and methane (CH_4), both of which are the direct by-products of OC mineralization. The extent to which flooded OC is mineralized in reservoirs, and the amount of MeHg produced, may depend on the amount and type of OC flooded (Kelly and others 1997). For example, the quantity of easily decomposed labile OC flooded may be important to short-term decomposition processes fuelling MeHg and GHG production, whereas the total quantity of OC flooded may affect the long-term duration of production. To begin examining the link between OC mineralization and Hg methylation in reservoirs, a whole-ecosystem experiment was initiated at the Experimental Lakes Area (ELA) in northwestern Ontario in 1990/91 (Kelly and others 1997). The ELA Reservoir Project (ELARP) flooded a wetland complex because it was thought to provide the worst-case scenario for MeHg and GHG production due to the large stores of OC in peat available for decomposition over the long-term. Within three weeks of flooding, yields of MeHg and carbon GHGs from the reservoirs increased dramatically (Kelly and others 1997). Continued background monitoring of dissolved MeHg, CO_2, and CH_4 in the wetland reservoir showed that rates of MeHg and GHG production are well above pre-flood levels nine years after the initial inundation of the wetland (St. Louis and others submitted).

The main objective of the research presented here was to determine if flooding smaller quantities of OC compared to the ELARP would result in less MeHg and GHG production in reservoirs. We addressed this objective by initiating a new whole-ecosystem flooding experiment at the ELA in 1997. The FLooded Upland Dynamics EXperiment (FLUDEX) flooded three upland boreal forest sites that varied in amounts of OC stored in soils and vegetation. The three upland reservoirs (Low C, Medium C, and High C), with OC storage of between 30 900 and 45 900 kg C ha^{-1}, contained between 60 and 27 times less OC than stored in the ELARP wetland reservoir (1.26×10^6 kg C ha^{-1}). Here we calculate Hg input-output budgets for three seasons of flooding (1999-2001) to determine net MeHg and THg yields from the three FLUDEX reservoirs. We also estimated the net production of MeHg in each reservoir by calculating annual changes in pools of MeHg stored in flooded soils, periphyton, zooplankton, and fish. The resulting MeHg production rates in the FLUDEX upland reservoirs are compared to methylation rates observed in the ELARP wetland reservoir.

Overall, there was an initial pulse of MeHg production in all FLUDEX reservoirs that lasted for 2 years, after which time net demethylation began to reduce the pools of MeHg in the reservoirs. Rates of methylation were generally related to the total amount of OC flooded to create the reservoirs. This study should assist hydroelectric utilities and government agencies in making informed decisions about selecting sites for future reservoir development to reduce MeHg contamination of the reservoir fisheries.

Methods

Description of FLUDEX reservoir sites

The ELA camp is located 50 km southeast of Kenora, Ontario, on the Precambrian Shield. The ELA experiences a cold temperate continental climate with 32-year mean July and January temperatures of 18.5 and –17.3°C, respectively. Mean annual wet deposition from 1969-2001 was 699 mm, with ~25% of this wet deposition falling as snow. Upland areas at the ELA ranged from open lichen-covered granite/gneiss rocks to shallow nutrient-poor acidic soils supporting jack pine (*Pinus banksiana*), black spruce (*Picea mariana*) and paper birch (*Betula papyrifera*) forest communities.

Three 17-year-old fire-regenerated upland forest sites differing in amounts of OC stored in vegetation and soils were chosen in 1997 to be experimentally flooded. The High C site had 45 900 kg C ha^{-1}, the majority of which was stored in trees (57%) and the fungal/humic (FH) layer of the soils (34%; Table 4-1). The 0.74 ha High C site had rapid but imperfect drainage, occasionally resulting in pools of standing water. There were two basic vegetation communities in the High C site: 1) a jack pine (*Pinus banksiana*) dominated forest with an understory of wetland plants such as Labrador tea (*Ledum groenlandicum*), *Sphagnum* spp. mosses, and leatherleaf (*Chamaedaphne calyculata*) shrubs growing in low-lying, wet soils, and 2) a drier upland area dominated by jack pine and *Polytrichum* spp. mosses (Table 4-1).

Table 4-1. Carbon stores (kg C ha^{-1}) in the FLUDEX sites prior to flooding.

	High C Site	Medium C Site	Low C site
Dominant vegetation (percent coverage)	*Pinus/Ledum/Sphagnum* (53%) *Pinus/Polytrichum* (47%)	*Pinus/Betula* (100%)	*Pinus/Vaccinium* (73%) *Polytrichum/Cladina* (22%) *Organic pillows* (5%)
Range of soil depth (cm)[1]	6.3 - 105.0	15.6 - 90.6	0 - 69.0
Range of forest floor depth (cm)[1,2]	1.0 - 37	3.5 - 13.0	2.0 - 7.5
	kg C ha^{-1}	kg C ha^{-1}	kg C ha^{-1}
Carbon in trees	26 210	27 600	19 570
in foliage[3]	1 970	2 730	1 770
in bark[3]	2 440	3 760	1 970
in wood[3]	21 800	21 110	15 830
Carbon in shrubs, herbs, and mosses[3]	1 350	130	200
Carbon in litter and fungal/humic layer[2]	15 400	5 700	8 700
Carbon mineral layer[1]	2 900	1 500	2 400
Total soil carbon (including litter)[1]	18 300	7 200	11 100
Total carbon in above ground vegetation[3]	27 560	27 730	19 770
Total carbon (kg C ha^{-1})	**45 860**	**34 930**	**30 870**

[1] (Venkiteswaran and others submitted)

[2] where forest floor exists

[3] (Matthews and others submitted)

The 0.50 ha Medium C site had better drainage than the High C site, and relatively complete coverage of dry soils ranging from 15 to 90 cm in depth (Table 4-1). A dense jack pine forest with tall birch (*Betula papyrifera*) and alder (*Alnus* spp.) shrubs, an understory of blueberry (*Vaccinium* spp.) shrubs, and an extensive groundcover of various mosses and herbs dominated the site. Approximately 80% of the total OC (34 900 kg C ha^{-1}) was stored in trees, whereas 16% was stored in the FH layer of soils (Table 4-1). The 0.63 ha Low C site was located on a dry ridge-top. Approximately 73% of the site had shallow soils supporting sparse stands of jack pine and birch with a blueberry shrub dominated understory. 22% of the site had areas of thin glacial till with lichens, mosses, blueberry shrubs, and exposed bedrock. 5% of the area was covered with lichens (*Cladina* spp.), mosses (*Polytrichum* spp., *Racomitrium microcarpon*), and grasses (*Poa* spp.) overlying <10 cm of organic deposits. The Low C site contained 30 900 kg C ha^{-1}, with 64% and 28% stored in trees and the FH soil layer, respectively (Table 4-1).

For comparison with the FLUDEX reservoirs, the ELARP wetland reservoir consisted of a 2.4 ha centre pond surrounded by a 14.4 ha peatland, dominated by black spruce (*Picea mariana)*, larch (*Larix laricina*), *Sphagnum* spp., and Labrador tea and leatherleaf shrubs (Dyck and Shay 1999). The vast majority of OC in the site was stored as peat (1.26 x 10^{6} kg C ha^{-1}). Less than 2% of the OC stored in the peatland was living vegetation and/or litter.

Table 4-2. Physical properties of the FLUDEX upland reservoirs.

	High C reservoir			Medium C reservoir			Low C reservoir		
Surface area (m^2)	7 358			4 966			6 271		
Direct runoff area (m^2)	47 842			7 334			929		
Reservoir watershed area (m^2)	55 200			12 300			7 200		
Volume (m^3) above flooded soils	6 870			4 270			7 120		
Volume (m^3) in flooded soils	1 810			1 810			800		
	1999[1]	2000[2]	2001[3]	1999[1]	2000[2]	2001[3]	1999[1]	2000[2]	2001[3]
Water renewal time (days)	11	8	9	10	6	6	8	7	7
Reservoir turnovers	9	14	11	10	17	18	11	14	15
Days of weir outflow	96	105	103	95	104	108	91	100	100
Days to fill	8	10	13	9	11	9	13	15	17

[1] 1999: Start of flooding: 22-Jun; Start of drawdown: 04- Oct; Days water pumped 104.

[2] 2000: Start of flooding: 30-May; Start of drawdown: 21- Sep; Days water pumped 115.

[3] 2001: Start of flooding: 29-May; Start of drawdown: 24-Sep; Days water pumped 117.

Upland reservoir construction

Upland reservoirs were constructed by building dikes along low-lying contours of the sites followed by flooding with water pumped from a nearby oligotrophic lake. In areas where flooding would not exceed 1 m depth, gravel dikes embedded with plastic sheeting were constructed. Wooden structural walls were constructed in areas where impoundment levels were to be greater than 1 m in depth. Sealing was achieved by incorporating a plywood-plastic-plywood "sandwich" technique on the walls, and a plastic and concrete grout seal at the base. Areas of sites exceeding the height of inundation were not diked, creating 'riparian zones' that were open to 4.7, 0.7 and 0.09 ha of upland above the High C, Medium C, and Low C reservoirs, respectively.

Beginning in 1999 (Table 4-2), water was pumped with a diesel pump into each reservoir from nearby oligotrophic Roddy Lake (L468) through aluminium irrigation pipe (Fig. 4-1). The reservoirs took between 8 and 17 days to fill to a maximum depth of 2 m and average depth of 1 m (Table 4-2). Water exited the reservoirs over v-notch weirs. In September/October, the reservoirs were emptied through gate valves installed at the bottom of the wooden walls to simulate drawdown in the shallow zones of northern hydroelectric reservoirs in the winter. Water volume and surface area of each reservoir was calculated using topographical maps determined from aerial photographs taken in 1982 and 1991.

Stores of MeHg and THg in the upland forest sites prior to flooding

Estimates of MeHg and THg stored in foliage, shrubs, ground cover, wood, and soils were calculated to determine the total mass of MeHg and THg in each reservoir prior to flooding. MeHg concentrations in foliage (birch and alder leaves and pine needles), shrubs (blueberry and Labrador tea leaves), mosses, lichens, and wood collected at the ELA in each site prior to flooding (Chapter 3, this thesis) were determined using cold vapour atomic fluorescence

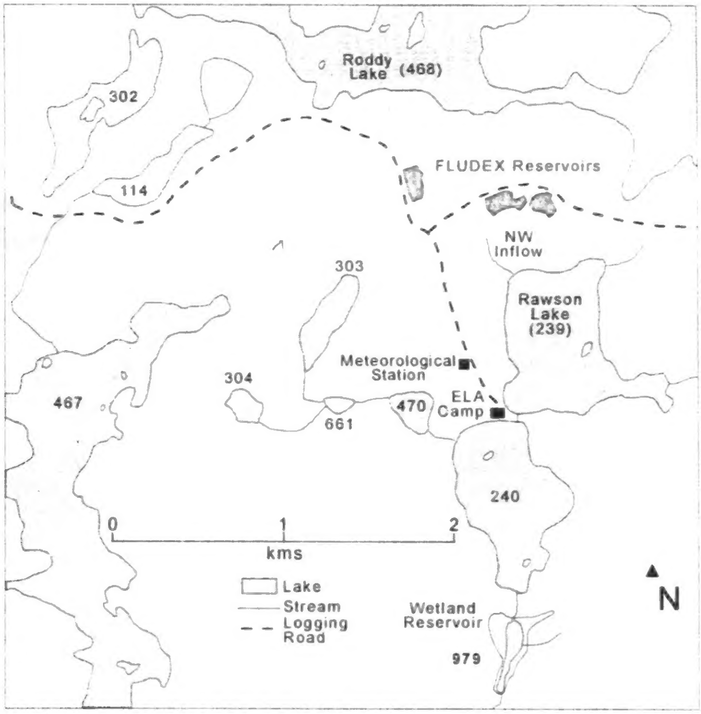
Roddy Lake (468)
302
114
FLUDEX Reservoirs
NW Inflow
303
Rawson Lake (239)
Meteorological Station
304
467
470
ELA Camp
661
240
0
1
2
kms
Lake
Stream
Logging Road
Wetland Reservoir
979
N

Figure 4-1: Location of study sites in relation to the Experimental Lakes Area field camp.

spectrophotometry (CVAFS) after overnight digestion in 25% KOH in methanol (Horvat and others 1993) and aqueous phase ethylation (Bloom 1989). THg concentrations were determined using CVAFS after digestion in 7:3 (vol:vol) nitric:sulphuric acid, as described in (Bloom and Fitzgerald 1988). National Research Council reference materials (lobster hepatopancreas and marine sediment) were regularly run on both the MeHg and THg analytical systems, the results generally indicating 80 - 110 % recovery of certified values.

The amount of foliage mass in the forest canopy in each reservoir was determined by weighing litterfall samples collected in duplicate at three sites within each reservoir (Matthews and others submitted). Two plastic containers (17 cm x 17 cm) were nested together; the bottom of the top container was removed, and a piece of 250 μm nitex mesh was wedged between them. Holes drilled in the bottom container allowed water to drain from the collectors. Collectors were attached to trees just above the water level and litter was collected from the containers monthly. Estimates of MeHg and THg stored in the canopy were calculated by multiplying MeHg and THg concentrations in foliage by the total dry weight of litterfall in year 2 when the forests in the reservoirs lost all foliage. MeHg and THg concentrations in other plant tissues were multiplied by the total dry weight of each tissue in shrubs, ground cover, and wood (Matthews and others submitted; Venkiteswaran and others submitted).

Storage of MeHg and THg in upland forest soils was calculated using concentrations measured in soil cores taken from random locations in each site in October 1998 (13, 14, and 11 cores in the High C, Medium C, and Low C sites, respectively). Cores were collected using a stainless steel barrel corer lined with plastic sleeves and were typically 8 to 15 cm long. Cores were sectioned into surficial litter/FH (the top 3-5 cm) and mineral layers, stored in acid-cleaned polypropylene cups, and immediately frozen. Cores were then lyophilized at –45 °C and homogenized using an acid cleaned mortar and pestle. THg and MeHg concentrations were determined at the University of Wisconsin-Madison and La Crosse Mercury Laboratories using the same techniques used to determine Hg concentrations in plants. The aerial mass of MeHg and THg stored in each soil layer was calculated by multiplying the MeHg or THg concentrations by a mean soil bulk density determined by drying the soils at 60°C (surficial FH density = 1.0, 0.5, and 0.5 g cm^{-2} and mineral soil density = 2.5, 3.0, and 2.0 g cm^{-2} in the High C, Medium C, and Low C sites).

Upland reservoir mercury input-output budgets

To calculate the *net* amount of MeHg or THg yielded from each experimental reservoir, quantities of MeHg and THg entering each reservoir were subtracted from quantities exiting (see St. Louis and others 1996; Kelly and others 1997). Input-output budgets were calculated for the period beginning on the first day that pumping began until the end of the drawdown (14-18 weeks each year; Table 4-2).

General Hg and Water Chemistry Sampling Methods and Analytical Techniques

All water samples taken to calculate MeHg and THg input-output budgets were collected in Teflon bottles using the clean-hands-dirty-hands sampling protocol (St. Louis and others 1996). MeHg water samples were frozen until analyzed. THg water samples were preserved using trace metal grade concentrated HCl to 1% of total sample volume. 20% of all samples were taken in duplicate. MeHg in water was analyzed by CVAFS after distillation (Horvat and others 1993) and aqueous phase ethylation (Bloom 1989) (detection limits = 0.01-0.02 ng L^{-1} at a blank level of 0.05-0.1 ng L^{-1}). Total Hg in water was analyzed using CVAFS as described in (Bloom and others 1988) with detection at 0.2-0.3 ng L^{-1} at a blank level of 0.3-0.4 ng L^{-1}. Flett Research Ltd. (Winnipeg, Manitoba, Canada) analyzed all water samples for MeHg and THg, with the exception of bottom water samples, which were analyzed at the University of Wisconsin Mercury Laboratories. Spike recoveries for MeHg and THg were generally >80% and >90 %, respectively.

Average MeHg and THg concentrations in water pumped in and out of each reservoir on two consecutive sampling dates were multiplied by the volume of water that entered each reservoir between the two dates to calculate mass inputs or outputs for that period. Inputs/outputs of MeHg and THg were calculated by summing the inputs/outputs for each sampling period throughout the period of flooding.

Surface water samples were taken concurrently with Hg water samples in Nalgene polypropylene bottles and analyzed at the ELA water chemistry laboratory for dissolved organic carbon (DOC) as in Stainton and others (1977). Oxygen concentrations and temperature in each reservoir were monitored weekly using an YSI oxygen/temperature probe. Temperatures were also measured continually in 2001 using Onset Hobo data loggers.

Inputs

Inputs of MeHg and THg to the upland reservoirs included water pumped from Roddy Lake, wet deposition in open areas, throughfall through the forest canopy, and direct runoff from the ungauged terrestrial areas above each reservoir (Appendix 8).

- Pumped inflow

Volume of water pumped into the reservoirs was regulated and monitored by means of control valves and in-line flow meters. Daily readings and adjustments were made in an attempt to equalize water residence times among the reservoirs (Table 4-2). Water at the end of the inflow pipe was sampled weekly for concentrations of MeHg and THg for the first month after flooding and then biweekly until reservoir drawdown.

- Wet deposition in the open and throughfall

Standard recording gauges at the ELA meteorological site were used to determine wet deposition volume in open areas. Wet deposition during the May to September study period was 13% (425 mm), 54% (580 mm), and 16% (399 mm) above the 32-year average (377 mm) average in 1999, 2000, and 2001, respectively. The volume of throughfall was determined using standard rain gauges placed under the forest canopy in the Medium C and Low C reservoirs. The areas of the reservoirs that received either direct wet deposition or throughfall were calculated from the % canopy cover at each site.

Wet deposition in the open was collected for MeHg and THg analysis at the ELA meteorological site in 1998-1999 into ultra clean wide mouth Teflon jars placed on acid washed plexiglass trays secured to wooden posts (St. Louis and others 2001). In 2000, wet deposition samples were collected in automated collectors installed on a cliff in the Lake 658 watershed ~7 km from the FLUDEX sites. Duplicate throughfall collectors, 1m in length, were set up at the sites by attaching wooden eavestrough holders to the trunk of trees. Within 15 minutes of the beginning of a precipitation event, acid-washed Teflon lined eavestroughing (12.5 cm wide by 75 m long) was set out on the holders. Throughfall drained into 1 L acid washed wide-mouth Teflon jars through acid washed nitex screen to remove large particles (St. Louis and others 2001). Throughfall was collected from all three sites prior to flooding in 1998, and after flooding from the Medium C reservoir in 1999, and the Low C reservoir in 2000 and 2001. There were no significant difference in MeHg and THg concentrations in throughfall among upland reservoirs when sampled concurrently in 1998 (ANOVA; $p=0.92$ and 0.39; $n=34$ and 20 for MeHg and THg, respectively; Appendix 9).

Wet deposition inputs of MeHg and THg (mg ha^{-1}) were estimated by multiplying the volume-weighted concentrations for each season by the total volume of either wet deposition in the open or in throughfall in the period that the reservoirs were flooded. Concentrations of MeHg in wet deposition in the open were not measured in 2001, so the volume-weighted concentrations from 2000 were used to estimate input for 2001.

- Direct runoff

The volume of water entering each reservoir in direct runoff through undiked areas was estimated on an aerial basis using a nearby gauged subcatchment with similar soils and vegetation (NW inflow to Rawson Lake, 56.4 ha; Fig. 4-1). However, because the NW inflow to Rawson Lake contained ~3% wetland and the presence of wetlands in catchments has been shown to affect concentrations of MeHg in runoff (St. Louis and others 1994), samples used to determine MeHg and THg concentrations in direct runoff were taken from the Lake 114 inflow ungauged weir site. The Lake 114 catchment contained no wetlands and was dominated by purely upland stands of jack pine and paper birch (*Betula papyrifera*) of similar post-fire age as the forests in and above the reservoirs. Runoff from the Lake 114 catchment was episodic during the summer months depending on rainfall intensity and antecedent moisture conditions. Samples were therefore taken opportunistically from May to September when there was runoff from this catchment. Inputs of MeHg and THg from direct runoff into the upland reservoirs were calculated by multiplying the estimated runoff water volume form the NW inflow by the average concentrations measured at the Lake 114 inflow weir.

Outputs

Mercury exited the upland reservoirs in water flowing over the weirs or out the drains during drawdown, in seepage, and as gaseous elemental Hg (Hg^0) fluxing off the surfaces of the reservoirs (Appendix 8).

- Weir outflow

Samples were collected for MeHg and THg analyses from above the reservoir outflow weirs weekly during the first month of flooding, and then biweekly until autumn drawdown.

- Seepage

Seepage was estimated in the water balance as the residual term and verified by independent seepage measurement surveys performed periodically during the study period. To estimate Hg loss due to seepage, we multiplied the seepage water volume by the concentration of MeHg and THg measured at the weir. Surface water concentrations were used because we could not confirm the origin of the seepage water and we assumed that Hg concentrations in the water flowing over the weir were representative of concentrations in the reservoir.

- Drawdown

The drawdown water volume was separated into two components: 1) surface waters estimated as 95% of the total water column volume and 2) bottom waters (1.5 cm from soil-water interface) estimated as 5% of total water column volume. To calculate the mass of MeHg and THg exiting through the drawdown pipe, the volume of surface water was multiplied by MeHg and THg concentrations in samples taken at the weir just prior to the beginning of drawdown, whereas the volume of bottom water was multiplied by average bottom water concentrations measured in samples taken 1-2 weeks prior to drawdown.

- Hg^0 fluxes from reservoir surfaces

An important loss of Hg out of aquatic ecosystems may be the flux of Hg^0 from the water surface to the atmosphere. Hg^0 can be formed as a result of the photoreduction of MeHg and HgII in the water column (Amyot and others 1994; Sellers and others 2001). Although we did not directly measure fluxes of Hg^0 from the reservoir surfaces, we determined the upper and lower limits of dissolved Hg^0 concentrations in the water column. In 2001, samples for dissolved Hg^0 analysis were collected by completely filling a 2.5 L acid-washed glass bottle with reservoir surface water. In the laboratory, 250 mLs of water was removed from each bottle to create a headspace. Bottles were fitted with caps containing an inlet consisting of one piece of 5 mm Teflon tubing that extended to the bottom of the bottle and an outlet attached to a gold-coated-bead Hg^0 trap. UHP nitrogen was then bubbled through the sample at a rate of ~5 L min^{-1} for between 2 and 6 h in the dark, forcing dissolved Hg^0 out of solution and onto the gold traps. Quantification of Hg^0 collected on the gold traps was performed in the ELA Hg clean laboratory with a Tekran Model 2500 Mercury

Vapour Detector (see Lindberg and others 2000), using dual gold-coated bead amalgamation with a detection limit of 0.5 pg.

Sources of error in input-output budgets and statistical analysis

The two main sources of error in our input-output budget were associated with the analyses of samples for MeHg and THg, and the estimation of water volumes used in our calculations. Analytical precision was ±0.02 ng L^{-1} for MeHg and ±5 ng L^{-1} for THg. These values were within 19% and 10% of a consensus value obtained in a recent inter-lab comparison. Standard procedures and equipment were used for the measurement of precipitation, flow (v-notch weirs and calibrated flow meters), and water levels (precision recorders and data loggers). Evaporation was measured directly with Class A evaporation pans placed in shallow areas of the reservoirs. While it is difficult to independently assess error associated with each of these parameters, errors generally accepted are 5% for precipitation, weir flow, and water levels, ~15% for evaporation, and ~18% when applying gauged runoff to similar ungauged areas (Winter 1981). By far, the major component of the water balance is the inflow by pumping where the associated errors were 5%.

The FLUDEX could not replicate experimental units due to the difficulty replicating whole-ecosystem experiments (Schindler 1998). However, data trends, as well as pre- and post-flood comparisons, convincingly demonstrate effects of flooding on changes in MeHg and THg concentrations among the reservoirs. Inferential statistics are used to emphasize results, acknowledging the implication of pseudoreplication (Hurlbert 1984).

Pools of MeHg stored in upland forest reservoirs

To estimate the total net production of MeHg in the reservoirs at the end of the each flooding season (September), MeHg stored in soils and food web organisms was calculated, and added to the net loss of MeHg out the outflow calculated using the input-output budgets. For each year, these total stores of MeHg were then subtracted from stores calculated in the previous September.

Soils

MeHg stored in soils in September was estimated from MeHg concentrations measured in cores collected using an acid-cleaned acrylic tube sampler from three sites located in each reservoir. Cores were sectioned into surficial litter/FH and mineral layers, stored in acid-cleaned polypropylene cups, and immediately frozen. MeHg masses were obtained using the same analytical methods use to obtain MeHg concentrations in soil cores collected prior to flooding (see section on MeHg and THg storage in unflooded upland forest sites).

Food web

Periphyton was collected from 2 m long x 2 cm diameter fir dowels that were hung among the trees at five stations within each reservoir (Appendices 9 - 11) prior to flooding in years 2 and 3 (2000-2001). Each September, periphyton was rinsed from two dowels at each station into plastic bags and immediately frozen. Periphyton was analyzed for MeHg at Flett Research Ltd. using the same analytical methods as for plant samples (see above section on MeHg stores in plants). The mass of MeHg in the periphyton community was calculated by multiplying MeHg concentrations with in the total mass of periphyton (unpublished data, D. Findlay, Freshwater Institute, Winnipeg, MB and J. Venkiteswaran and S. Schiff, University of Waterloo, Waterloo, ON) in each reservoir. Dowels were not installed in the first year of flooding (1999), but grab samples of periphyton attached to trees in each reservoir were collected in September and analyzed for MeHg concentrations. Because quantitative estimates of periphyton biomass were not available, year 1 periphyton MeHg mass was calculated by multiplying the concentration of grab samples taken in 1999 with the estimate of biomass in year 2 (2000).

Zooplankton were collected for MeHg analysis using a 150 μm sweep net from the open regions of each reservoir and immediately frozen in whirl-pak bags (Paterson and others 1998). Flett Research Ltd. analyzed zooplankton for MeHg using the same methods outlined for plants and periphyton. Zooplankton biomass was determined by collecting samples at 10-12 stations within each reservoir using a quantitative tube sampler (Paterson and others 1997; Peech Cherewyk 2002). MeHg pools were obtained by multiplying areal zooplankton biomass in each reservoir by the MeHg concentrations in zooplankton. Biomass and MeHg concentrations in benthic organisms were not measured.

Fine scale dace (*Phoxinus neogaeus*) were introduced annually into the reservoirs at densities between 0.35 and 0.44 fish m^{-2}. Fish were caught each September with either minnow traps (1999) or small mesh gill nets (2000 and 2001). Approximately 0.2 g of white epaxial muscle were analyzed for THg using cold vapour atomic absorption spectrophotometry (Hendzel and Jamieson 1976) in the Freshwater Institute Mercury Laboratory (Winnipeg, MB). The majority of THg in fish is MeHg (Hall and others 1997; Bodaly and Fudge 1999). Muscle THg concentrations are ~30% higher than whole body concentrations, therefore a conversion factor of 0.7 was applied to muscle THg concentrations to calculate total body burdens. To determine THg stored in fish in each reservoir, average body burdens at the beginning of each season were subtracted from final average body burdens and multiplied by an estimate of fish biomass.

Determination of methylation and demethylation rates

Whole-reservoir rates of methylation or demethylation were estimated by dividing the net change in MeHg stored at the end of one flooding season (September) by the number of days that the reservoirs were flooded each year. For the first year of flooding, we subtracted the MeHg stored in vegetation and soils prior to flooding from the total MeHg stored in the reservoirs in September 1999.

Results and Discussion

MeHg and THg storage in unflooded upland forest sites

Prior to flooding, average MeHg concentrations in plants and soils ranged from 0.12 to 1.13 ng g^{-1} (Table 4-3). Average THg concentrations were between 3.6 and 96.0 ng g^{-1}. MeHg stores prior to flooding in the High C site (178 mg ha^{-1}) were 5-6 fold greater than in the Medium C and Low C sites (37 and 39 mg ha^{-1}, respectively; Table 4-3). THg stored in vegetation and soils in the High C and Medium C sites (14 900 and 14 500 mg ha^{-1}) were similar, and about 2 fold greater than in the Low C site (7 580 mg ha^{-1}; Table 4-3). For all sites, MeHg and THg were predominantly stored in soils (81-95% and 95-98%, respectively). MeHg was primarily found in the FH layer of soils in the High C and Low C sites, whereas MeHg mass was split evenly between the FH and mineral layers of soils in the Medium C site (Table 4-3). In the Medium C and Low C sites, THg masses were 2 times higher in the mineral layers than in the overlying FH layer. In the High C site, storage of THg in the FH layer was greater than the mineral layer.

The large difference between stores of MeHg and THg at our sites is most likely due to the higher deposition of THg than MeHg to ELA forests over the 18 years of fire regeneration (St. Louis and others 2001). The amount of THg present as MeHg (%MeHg) in the sites prior to flooding was greater in the High

Table 4-3. Average concentrations (ng g^{-1}) and stores (mg ha^{-1}) of methylmercury (MeHg) and total mercury (THg) in the FLUDEX sites prior to flooding.

	High C				Medium C				Low C			
	MeHg		THg		MeHg		THg		MeHg		THg	
	ng g^{-1}	mg ha^{-1}	ng g^{-1}	mg ha^{-1}	ng g^{-1}	mg ha^{-1}	ng g^{-1}	mg ha^{-1}	ng g^{-1}	mg ha^{-1}	ng g^{-1}	mg ha^{-1}
Above ground												
wood and bark	0.23	21	3.6	330	0.23	15	3.6	230	0.23	13	3.6	210
shrubs and ground cover	0.50	2	37.8	290	0.49	<1	43.5	10	0.62	<1	49.4	20
foliage	0.12	1	11.3	100	0.12	1	11.3	90	0.12	1	11.3	70
Fungal/humic soil layer	1.13	113	89.1	8 960	0.20	10	44.2	4 450	0.52	16	39.2	2 360
Mineral soil layer	0.17	41	51.8	5 200	0.04	11	96.0	9 670	0.08	9	81.0	4 920
Total		*178*		*14 880*		*37*		*14 450*		*39*		*7 580*

C site (1.1%) compared to the Medium C and Low C sites (0.2% and 0.4%, respectively) likely due to the presence of pockets of saturated wetlands in the High C site, where methylation is known to occur (St. Louis and others 1994; Branfireun and others 1998). Over the long term, litterfall represents an important input of both OC and Hg to growing forests (St. Louis and others 2001). However, in the FLUDEX reservoirs, litterfall represented a one-time pulse of OC from the canopy into our reservoirs. This OC addition provided additional substrates for decomposition in the second year of flooding after the trees had died (Matthews and others submitted; Appendix 11). However, despite the large flux of litterfall into the reservoirs in 2000, the input of litterfall MeHg (1.1-1.7 mg ha^{-1} season^{-1}) was very low due to extremely low concentrations of MeHg in litter. THg inputs were ranged from 65 to 422 mg ha^{-1}.

Water and mercury budgets

Inputs

- Pumped input

Concentrations of Hg in water entering each reservoir were always between 0.007 and 0.151 ng MeHg L^{-1} and between 0.80 and 2.25 ng THg L^{-1}. Pumped inflow over the course of each season contributed on average 95±1% of total water inputs to all three upland reservoirs (Table 4-4). Inflow water contributed between 3 - 9 mg MeHg (Table 4-5) and 77 - 150 mg THg (Table 4-6) among reservoirs over the three-year study. This input was equivalent to 87% - 98% of total MeHg inputs and 47% - 91% of all THg inputs to the upland reservoirs.

- Wet deposition and throughfall

Concentrations of Hg in wet deposition collected in the open in 1998-2000 ranged from 0.007 - 0.228 ng MeHg L^{-1} and 0.21 - 25.55 ng THg L^{-1} (Appendix 14). Concentrations of Hg in throughfall ranged between 0.062 - 0.344 ng MeHg L^{-1} and 3.10 - 33.36 ng THg L^{-1}. Volume weighted seasonal averages of MeHg

Table 4-4. Water inputs and outputs (m^3) to and from the FLUDEX upland reservoirs in 1999, 2000, and 2001.

	High C reservoir			Medium C reservoir			Low C reservoir		
	Year 1	Year 2	Year 3	Year 1	Year 2	Year 3	Year 1	Year 2	Year 3
Inputs									
Inflow	78 600	93 300	84 800	76 700	69 600	74 700	96 900	108 600	106 900
Wet deposition in the open	1 220	1 790	1 310	820	1 200	880	1 140	1 680	1 230
Throughfall	710	990	1 300	480	670	880	560	780	1 020
Direct run-off	4 680	12 220	4 950	710	1 850	780	60	150	90
Total water input	*85 210*	*108 300*	*92 360*	*78 710*	*73 320*	*77 240*	*98 660*	*111 210*	*109 240*
Outputs									
Weir	60 200	80 200	64 000	60 800	57 200	63 700	33 500	42 500	51 500
Seepage	16 000	15 900	13 000	15 400	13 900	14 500	23 100	19 100	12 200
Bedrock fracture							36 900	43 400	40 800
Outflow pipe (total)									
Surface water	6 530	6 530	6 530	4 050	4 050	4 050	6 760	6 760	6 760
Bottom water	340	340	340	210	210	210	360	360	360
Evaporation	1 620	1 800	1 900	1 350	1 300	1 350	1 380	1 650	1 570
Canopy interception	840	1 280	370	570	870	250	660	1 000	290
Total water output	*85 530*	*106 050*	*86 140*	*82 380*	*7 7530*	*84 060*	*10 2660*	*114 770*	*113 480*

Table 4-5. Inputs and outputs of methylmercury (mg MeHg) and MeHg exports (mg ha^{-1}) to and from FLUDEX upland reservoirs.

	High C reservoir			Medium C reservoir			Low C reservoir		
	Year 1	Year 2	Year 3	Year 1	Year 2	Year 3	Year 1	Year 2	Year 3
Inputs									
Inflow	3.4	4.3	7.2	3.4	3.0	6.0	4.1	5.1	9.1
Wet deposition in the open	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Throughfall	0.2	0.2	0.4	0.1	0.1	0.3	0.1	0.1	0.3
Direct run-off	0.2	0.3	0.5	<0.1	0.1	0.1	<0.1	<0.1	<0.1
Total MeHg input	*3.9*	*4.9*	*8.2*	*3.6*	*3.2*	*6.6*	*4.4*	*5.2*	*9.4*
Outputs									
Weir	39.4	48.5	31.4	52.0	49.1	40.0	14.9	18.1	13.2
Seepage	9.3	9.2	6.3	11.6	11.8	9.0	9.7	7.4	3.4
Bedrock fracture							15.5	16.7	11.2
Outflow pipe									
Surface water	3.1	2.9	2.7	2.6	7.3	1.4	1.7	1.4	1.3
Bottom water	0.6	0.9	0.2	0.2	0.1	0.1	0.2	0.1	0.1
Total MeHg output	*52.4*	*61.5*	*40.6*	*66.4*	*68.3*	*50.4*	*42.0*	*43.6*	*29.2*
MeHg output-MeHg input	*48.5*	*56.6*	*32.4*	*62.8*	*65.1*	*43.8*	*37.6*	*38.4*	*19.7*
MeHg yield[1] (mg MeHg ha^{-1})	**65.9**	**77.0**	**44.0**	**126**	**131**	**88.2**	**59.9**	**61.2**	**31.5**

[1] MeHg yield = MeHg output - MeHg input / reservoir area (see Table 4-2)

Table 4-6. Inputs and outputs of total mercury (mg THg) and THg (mg ha^{-1}) exports to and from the FLUDEX upland reservoirs.

	High C reservoir			Medium C reservoir			Low C reservoir		
	Year 1	Year 2	Year 3	Year 1	Year 2	Year 3	Year 1	Year 2	Year 3
Inputs									
Inflow	78.2	125.4	89.8	76.6	92.1	79.0	96.3	147.1	113.0
Wet deposition in the open	16.1	6.9	12.1	10.8	4.6	8.1	15.1	6.5	11.4
Throughfall	17.6	7.7	24.9	12.0	5.2	16.9	13.8	6.0	19.5
Direct run-off	42.6	124.8	41.4	6.4	18.7	6.5	0.5	1.6	0.8
Total THg input	*155*	*265*	*168*	*106*	*121*	*111*	*126*	*161*	*145*
Outputs									
Weir	186	256	181	258	196	159	87.1	91.0	86.4
Seepage	48.0	53.3	37.2	74.6	53.4	37.2	76.6	44.0	22.6
Bedrock fracture							123	101	74.7
Outflow pipe									
Surface water	14.4	21.4	11.8	7.9	10.2	5.7	9.5	13.1	7.2
Bottom water	4.6	0.9	0.9	0.7	0.4	0.4	1.2	0.8	0.4
Total THg output	*253*	*332*	*230*	*341*	*260*	*203*	*297*	*250*	*191*
THg output-THg input	*97.9*	*67.1*	*62.2*	*235*	*139*	*92.0*	*171*	*88.7*	*46.6*
THg yield[1] (mg THg ha^{-1})	**133**	**91.2**	**84.5**	**474**	**280**	**185**	**273**	**141**	**74.3**

[1] THg yield = THg output - THg input / reservoir area (see Table 4-2)

and THg in wet deposition in the open (0.07±0.02 - 0.13±0.02 and 3.87±0.72 - 13.21±2.71 ng L^{-1}, for MeHg and THg respectively) were always lower than those in throughfall (0.15±0.04 - 0.3 ± 0.02 and 7.71±2.46 - 24.70±4.81 ng L^{-1}; Appendix 15).

In the first two years of flooding, throughfall volume was 46% and 44% of wet deposition. By the third year of flooding, when most foliage had fallen off the dead trees, 78% of wet deposition made it through the canopy. Water inputs from wet deposition in the open and throughfall only accounted for between 1% and 2% of total water inputs (Table 4-4). MeHg inputs from wet deposition in the open and throughfall were low (<0.5 mg), constituting 4% - 8% of total MeHg inputs, whereas THg inputs in wet deposition and throughfall ranged between 10 - 37 mg, contributing 6% - 23% of all THg inputs (Table 4-5).

- Direct runoff

Concentrations of Hg in runoff from the L114 upland catchment ranged between 0.013 - 0.127 ng MeHg L^{-1} and 6.56 - 14.93 ng THg L^{-1} (Appendix 15). The volume of water entering the High C reservoir in runoff was estimated to vary between 5% and 11% of the total water inputs among years (Table 4-4). MeHg inputs due to runoff into the High C reservoir were <1 mg for the flooding season, or 5% - 6% of total MeHg inputs (Table 4-5). THg inputs to the High C reservoir from direct runoff ranged from 41 – 125 mg and contributed up to 47% of all THg inputs (Table 4-6). Total runoff water inputs to the Medium C and Low C reservoirs were <3% of total water inputs, delivering less than 3% of the total MeHg inputs. Direct runoff contributed from <1% to 16% of all THg inputs to the Medium C and Low C reservoirs (Table 4-6).

Outputs

- Weir outflow

Average MeHg concentrations in outflow waters from all three upland reservoirs were ~3.4 times greater than concentrations in inflow water within the first week of flooding. Concentrations of MeHg in outflow water from all reservoirs remained elevated for the duration of the experiment, peaking at 1.60 ng L^{-1} in year 2 in the Medium C reservoir (Fig. 4-2A). MeHg concentrations in the Medium C reservoir were almost always higher than in the High C and Low C reservoirs until just prior to drawdown (Fig. 4-2A). Concentrations of THg in water exiting over the reservoir weirs were much higher than in inflow water (Fig. 4-2B). Concentrations of THg in outflow water tended to decrease throughout each season, and average annual mean THg concentrations in all reservoirs decreased over the first three years of flooding.

As in the FLUDEX upland reservoirs, concentrations of MeHg in the outflow waters of the ELARP wetland reservoir increased within the first month of flooding and remained elevated over the course of the three-year period, reaching a maximum outflow concentration of 3.2 ng L^{-1} in year 3 post-flood (Fig. 4-2A). Average annual MeHg concentrations in the wetland reservoir increased over the first three years of flooding, while THg concentrations stayed relatively constant.

Although water concentrations are important in determining MeHg accumulation in aquatic organisms (Paterson and others 1998), they cannot be used to examine relative rates of MeHg production because the flushing rates were different among reservoirs. Differences in MeHg and THg cycling due to different flushing rates were eliminated by using Hg input-output budgets. Surface water yields (measure outflow/total inflow) averaged 71%, 79%, and 40% for the High C, Medium C, and Low C reservoirs, respectively (Table 4-4). The lower surface flow yields for the Low C reservoir were due to uncontrollable flow through a subsurface bedrock fracture (see section on seepage). The mass

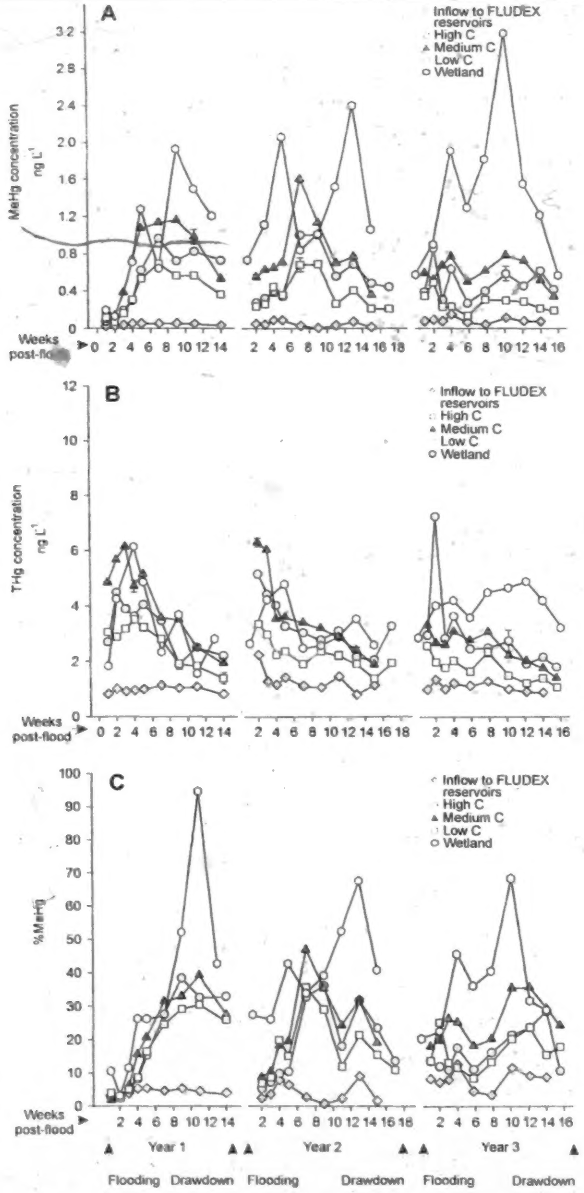

Figure 4-2. A: Concentrations of methylmercury (ng MeHg L^{-1}) in water flowing over the outflow weirs. B: Concentrations of total mercury (ng THg L^{-1}) in water flowing over the outflow weirs. C. The proportion of THg present as MeHg in the upland and wetland reservoirs (%MeHg). For A, B, and C: Error bars represent one standard error in duplicate samples. Differences in concentrations cannot be directly compared due to differences in reservoir flushing rates.

of MeHg and THg in water exiting the Medium C outflow weir (range = 40 - 52 mg MeHg and 160 - 258 mg THg) exceeded that leaving the High C and Low C reservoir outflow weirs (range = 13 - 49 mg MeHg and 86 - 256 mg THg) over the three-year period. Between 29% - 79% of the total mass of MeHg and THg exiting the reservoirs went over the weirs (Table 4-5 and 6).

An increase in the %MeHg often indicates increased net methylation rates within aquatic systems (Kelly and others 1995; Rudd 1995; Gilmour and others 1998). The %MeHg in FLUDEX reservoir outflows began to increase after the third week of flooding, and continued to increase peaking at over 45% MeHg midway through the second flooding season (Fig. 4-2C). The Medium C reservoir had higher % MeHg values than the two other upland reservoirs. The % MeHg decreased by the end of the second season of flooding, but began to increase again in the latter part of the third year of flooding. The % MeHg was generally higher in water exiting the ELARP wetland reservoir than in water exiting the FLUDEX upland reservoirs.

- Seepage

Between 11%-23% of the water inputs to the reservoirs seeped through the wooden and gravel dikes at the soil-bedrock interface. Seepage losses were similar among all reservoirs and study seasons. Seepage also occurred from a bedrock fracture in the Low C reservoir (~36% of total water inputs). Seepage rates decreased over time as dikes and wooden walls swelled and self-sealed. The total mass of MeHg and THg in seepage from the upland reservoirs was greatest in the Low C reservoir in the first year of flooding (25 mg MeHg and 199 mg THg; Table 4-5 and 6) because dike seepage volumes were highest in the Low C reservoir (Table 4-4) compared to the other upland reservoirs. Mass loss from seepage in the High C and Medium C reservoirs ranged from 6 - 12 mg MeHg and 37 - 75 mg THg over the three-year study.

- Drawdown

The average volume of water exiting the upland reservoirs through the drawdown pipe in autumn accounted for 6±0.4% of total water losses (Table 4-4). Concentrations of MeHg and THg in bottom waters ranged between 0.19±0.03 - 2.66±2.03 ng MeHg L^{-1} and 1.13±0.35 - 13.29±9.46 ng THg L^{-1} in all reservoirs over the three-year study (Appendix 16). The outputs of MeHg and THg through the drawdown pipes were similar among the upland reservoirs in all years, ranging from 3 - 11% of total MeHg outputs and 3 - 8% of all THg outputs.

- Hg^0 fluxes from reservoir surfaces

Average dissolved Hg^0 concentrations in samples taken in year 3 were higher in the Low C reservoir (65±9.6 pg L^{-1}) than the Medium C and High C reservoirs (36±7.6 pg L^{-1} and 22±6.3, respectively). These concentrations constituted <1%, 1.5%, and 3.7% of average seasonal THg concentrations in water in the High C, Medium C, and Low C reservoirs, respectively. Average dissolved Hg^0 concentrations were negatively correlated with average DOC concentrations ($r^2=0.9997$; Fig. 4-3A) and this negative relationship likely reflected less reduction of HgII to Hg^0 due to DOC screening of photoreducing UV energy (Scully and Lean 1994; Amyot and others 1997).

It is also possible that increased DOC concentrations contributed to HgII stabilization making it unavailable for reduction and therefore reducing Hg^0 evasion (unpublished data, K. Rolfhus). Concentrations of dissolved Hg^0 were only measured in year 3 when the annual average DOC concentrations were at their lowest (Fig. 4-3B), resulting in decreased UV screening compared to the first two years of flooding when average DOC concentrations were higher. Therefore, we expect that dissolved Hg^0 concentrations were very low in years 1 and 2. Due to the very small proportion of dissolved Hg^0 compared to water THg

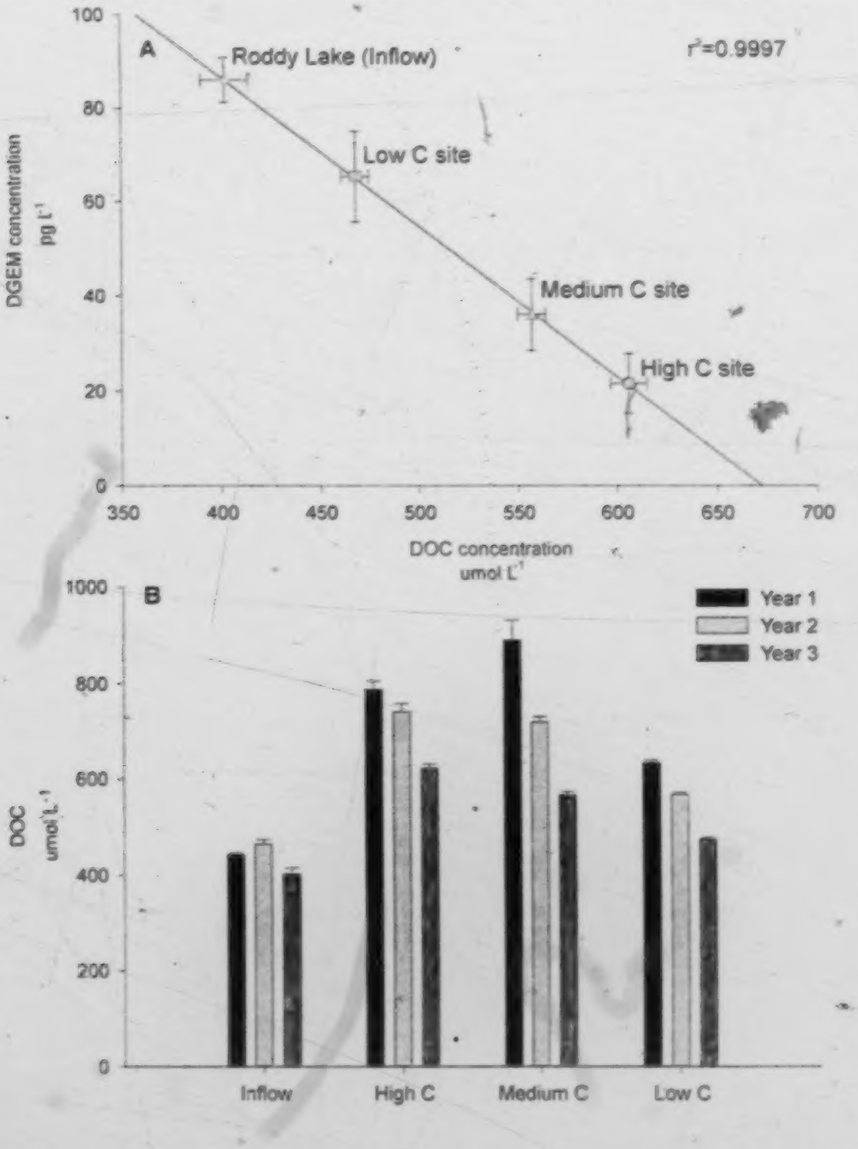

Figure 4-3. A: Year 3 concentrations of dissolved gaseous elemental mercury (DGEM) in relation to concentrations of dissolved organic carbon (DOC) in the reservoirs and source lake. B: Average seasonal DOC concentrations (μmol L^{-1}) in upland reservoirs.

concentrations, dissolved Hg^0 fluxes from the upland reservoirs were assumed to be insignificant and were not included in the mercury input-output budgets.

Mercury Yields

Net yields of MeHg and THg could not be calculated for the upland sites prior to flooding because the sites were not hydrologically gauged. However, a previous study at the ELA examined net yields of MeHg and THg from unflooded upland forested and wetland catchments over a three-year period (1990-1993) (St. Louis and others 1996). The purely upland catchment (Lake 114 inflow) retained both MeHg and THg (0.3 and 30 mg ha^{-1} yr^{-1} for MeHg and THg respectively; Table 4-7), and as a result, we assumed that the FLUDEX upland sites were also sinks for atmospheric inputs of MeHg and THg prior to flooding.

The hydrological control over the FLUDEX reservoir inflow and outflow rates enabled us to account for 98% of the water volumes over the three-year period (Table 4-4). All three upland reservoirs exported MeHg and THg during the three years of flooding. Net seasonal losses of MeHg from the Medium C reservoir exceeded those from the other upland reservoirs in all years, ranging from 0.58 to 1.22 mg MeHg ha^{-1} day^{-1} (Table 4-5 and 6, Fig. 4-4). Exports of MeHg from the High C and Low C reservoirs ranged from 0.27 to 0.67 mg ha^{-1} day^{-1} (Fig. 4-4). THg yields were also highest from the Medium C reservoir (1.58 – 4.55 mg ha^{-1} day^{-1}). THg exports from the High C and Low C reservoirs ranged between 0.63 to 2.63 mg THg ha^{-1} day^{-1} (Fig. 4-4). Net yields of both MeHg and THg, however, were highest in the first two years of flooding and decreased substantially in year 3.

Pools of MeHg stored in the reservoirs

Soils

Between 79% and 97% of the total MeHg stores in the reservoirs after

Table 4-7. Net yields of methylmercury (MeHg) and total mercury (THg) from unflooded upland forested and wetland catchments, and upland and wetland reservoirs at the Experimental Lakes Area. Negative values represent Hg sinks, positive values represent Hg sources. na= not available.

	MeHg Yield		THg Yield		Reference
	mg ha^{-1}yr^{-1}	mg ha^{-1}day^{-1}	mg ha^{-1}yr^{-1}	mg ha^{-1}day^{-1}	
Upland forests	-0.3	-0.0008	-30	-0.8	St. Louis and others (1996)
Catchments containing wetlands	-0.6	-0.002	-25	-0.07	St. Louis and others (1996)
Oligotrophic lake (L240)	0.04 0.06	0.0001 0.002	3.1	0.008	Sellers and others (2001) St. Louis and others (1996)
Upland reservoirs (FLUDEX)	NA[1]	0.27-1.22	na[1]	0.54-3.07	this study
Wetland reservoir (first 3 years of flooding)	19.9-69.8	0.05-0.15	3.1-130.3	0.008-0.36	St. Louis and others (submitted)

[1] Exports from upland reservoirs were only measured from May/June to September/October each year

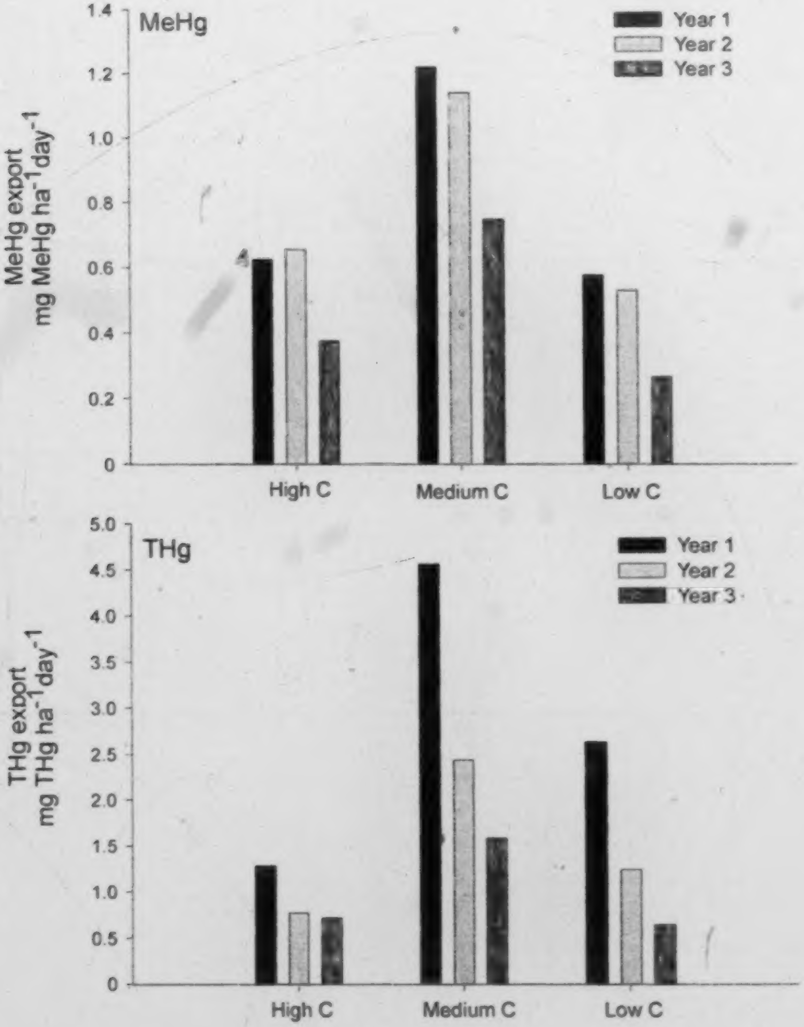

Figure 4-4. Export yields of methylmercury (mg MeHg ha^{-1} day^{-1}) and total mercury (mg THg ha^{-1} day^{-1}) from the upland reservoirs.

inundation were found in the litter/FH and mineral soils (Table 4-8). Post flood MeHg stores were generally higher in the litter/FH layer of soils (between 200 and 1700 mg ha^{-1}) than in the mineral soils (between 30 and 1500 mg ha^{-1}). Total MeHg stores in both the litter/FH and mineral layers combined were always highest in the High C reservoir and lowest in the Low C reservoir (Table 4-8). In all FLUDEX upland reservoirs, flooding resulted in a large increase in MeHg mass stored in soils. MeHg stores in soils over three years of flooding increased 9 - 21, 37 - 70, and 9 - 25 times those found in soils prior to flooding in the High C, Medium C, and Low C reservoirs, respectively (Table 4-3 and 8). These large increases in MeHg stores in soils post flood suggest that flooded soils were the main sites of MeHg production. The biomethylation of inorganic Hg depends on the metabolism of the methylating organisms (most likely sulphate reducing bacteria; Compeau and Bartha 1985) and anoxic soils would likely provide a favourable environment for growth of these organisms (Gilmour and Henry 1991). MeHg pools in soils decreased in the third year of flooding, but not to levels seen prior to inundation (Fig. 4-5). Similar conclusions were reached in the first three years of flooding the ELARP wetland reservoir (Kelly and others 1997; St. Louis and others submitted).

Food web

The periphyton, zooplankton, and fish communities represented the smallest MeHg pools in the upland reservoirs at the end of each flooding season, accounting for 1% to 10% of total MeHg stores (Table 4-8). MeHg stores were less than 15 and 6 mg ha^{-1} in periphyton and zooplankton, respectively and between 13 and 50 mg ha^{-1}in fish. Pools of MeHg in food web organisms generally did not differ among reservoirs. Stores of Hg in fish generally increased over time each year of flooding (Table 4-8), whereas MeHg stores in zooplankton decreased after the first season of flooding. There were no temporal patterns

Table 4-8. Methylmercury (mg MeHg ha^{-1}) stored in soils, vegetation, and biota in upland forest reservoirs and exported over the weir.

	Soils		Food web							
Reservoir	Fungal/ humic	Mineral	Periphyton[1]	Zooplankton	Fish	Yield over weir	Total MeHg	% of total MeHg in soils	% of total MeHg in food web	% of total MeHg yielded
High C										
Year 1 (1999)	807	621	6	0.4	13	66	1 510	94	1	4
Year 2 (2000)	1 72	1 510	10	<0.1	26	77	3 340	97	1	2
Year 3 (2001)	1 010	807	16	0.1	32	44	1 910	95	2	2
Medium C										
Year 1	487	704	20	5.1	19	127	1 360	87	3	9
Year 2	817	658	9	<0.1	35	131	1 650	89	3	8
Year 3	685	85	24	0.2	50	88	932	83	8	9
Low C										
Year 1	266	219	17	5.9	20	60	588	83	7	10
Year 2	564	51	11	<0.1	33	61	721	85	6	9
Year 3	202	29	7	1.3	26	32	297	78	11	11

[1] Because quantitative periphyton biomass was not obtained in year 1, year 2 MeHg biomass data were used to calculate year 1 MeHg pools (see text)

observed in MeHg pools in periphyton.

Total MeHg production in upland forest reservoirs

The total storage of MeHg in our upland reservoirs ranged from 290 mg ha^{-1} to over 3300 mg ha^{-1} (Table 4-8). Within each year, MeHg storage in the High C reservoir (1500-3300 mg ha^{-1}) was up to 2 times higher than storage in the Medium C reservoir (910-1600 mg ha^{-1}) and 3 - 7 times higher than in the Low C reservoir (290 - 710 mg ha^{-1}). MeHg stores within each reservoir were highest at the end of the second season of flooding, and then declined dramatically by the end of the third flooding season. However, total stores of MeHg in the reservoirs never approached the low levels observed prior to flooding (Fig. 4-5). Year 3 storage in the High C, Medium C, and Low C reservoirs were 12, 35, and 19 times higher than pre-flood storage, respectively (Fig. 4-5).

Overall, there was an initial pulse of MeHg production in all three reservoirs that lasted for 2 years, after which net demethylation began to reduce the pools of MeHg in all of our upland reservoirs. Rates of methylation and demethylation were generally related to the amount of total C stored in the reservoirs prior to flooding (Fig. 4-6). Rates of MeHg production were highest in the High C reservoir, producing 1280 and 1590 ng m^{-2} day^{-1} in the first and second years of flooding, respectively (Fig. 4-5). The rate of methylation in the Medium C reservoir in the first year of flooding (1270 ng m^{-2} day^{-1}) was very similar to that observed in the High C reservoir. The methylation rate in the Low C reservoir in the first year of flooding (530 ng m^{-2} day^{-1}) was more than 2.4 times lower than rates observed in the High C and Medium C reservoirs (Fig. 4-5). By the end of the second year of flooding, MeHg stores and rates of net methylation in the High C reservoir exceeded those in the Medium C (250 ng m^{-2} day^{-1}) and Low C (120 ng m^{-2} day^{-1}) reservoirs.

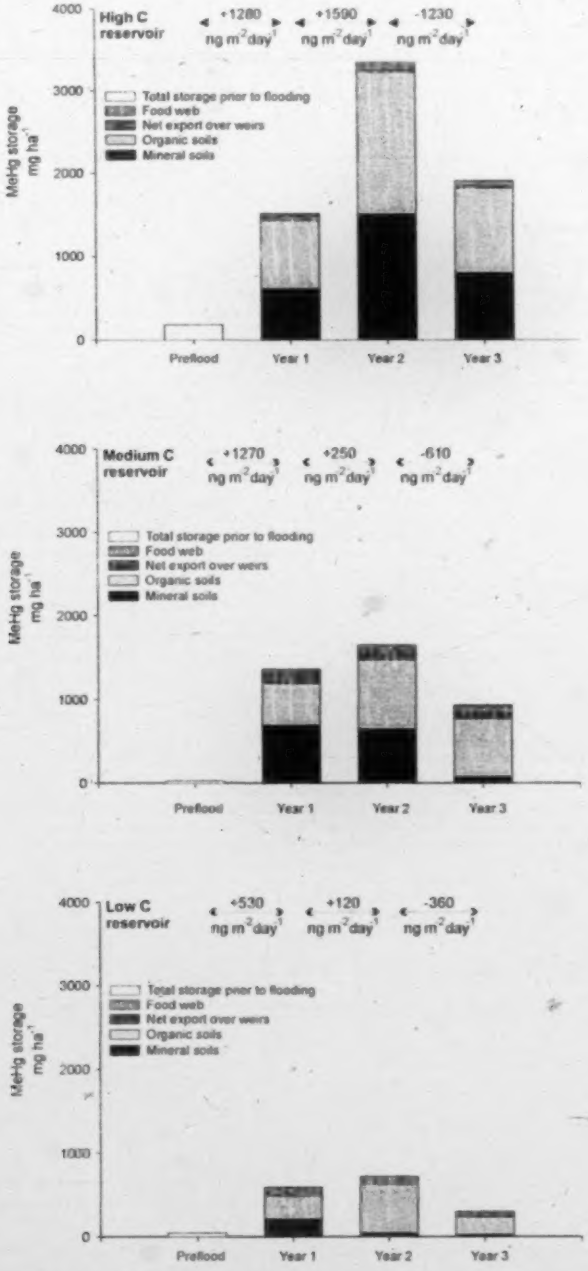

Figure 4-5. Methylmercury stores (mg MeHg ha^{-1}) in upland reservoirs. Methylation and demethylation rates are shown at the top of each panel; positive values represent net methylation and negative values represent net demethylation.

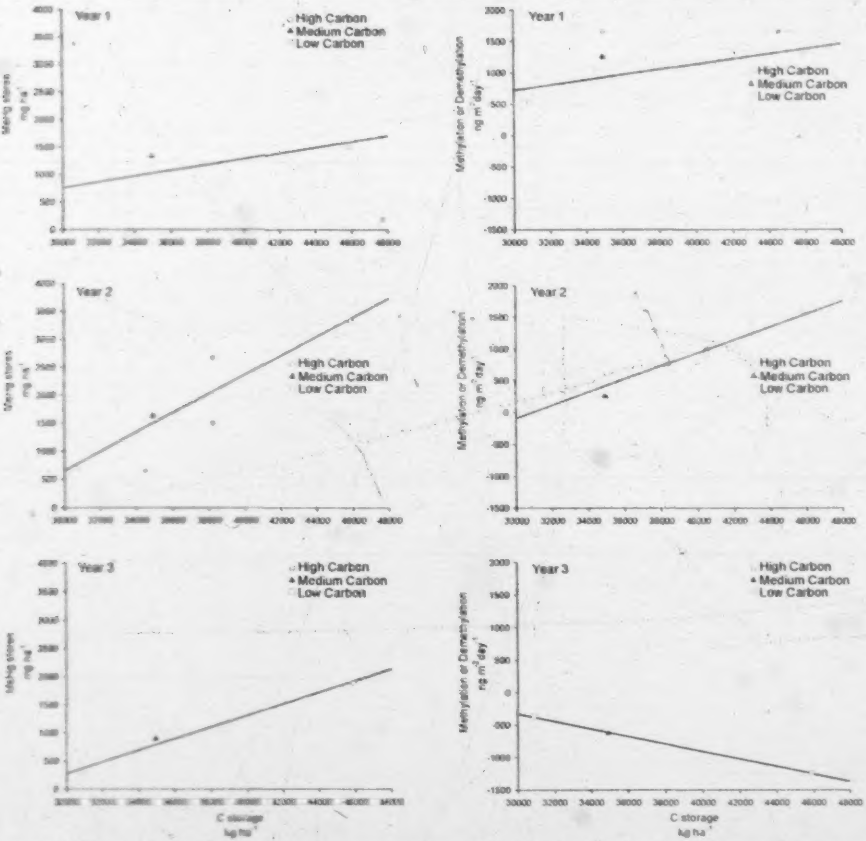

Figure 4-6. A: Methylmercury stores (mg MeHg ha^{-1}) in the upland reservoirs as a function of total pre-flood carbon stores (kg ha^{-1}). B: Methylation and demethylation rates (ng M^{-2} day^{-1}) in the upland reservoirs as a function of pre-flood carbon stores (kg ha^{-1}).

Net demethylation was observed in all three reservoirs in the third year of flooding (Fig. 4-6). Rates of net demethylation in the High C reservoir (1230 ng m^{-2} day^{-1}) exceeded those in the Medium C and Low C reservoirs (Fig. 4-6). Compared to the High C reservoir, net demethylation was 2 times lower in the Medium C reservoir (610 ng m^{-2} day^{-1}) and almost 3.4 times lower in the Low C reservoir (360 ng m^{-2} day^{-1}). We conclude that the destruction of MeHg was by microbial demethylation because photo-demethylation in the water column was minimal (see above), and the largest decreases in MeHg pools were observed in the litter/FH and mineral soil layers, where the microbial demethylators would expect to be most active (Robinson and Tuovinen 1984; Ullrich and others 2001).

Despite higher MeHg production in the High C reservoir, MeHg concentrations in water were higher in the Medium C reservoir compared to the other upland reservoirs, suggesting that there is a disconnect in the movement of MeHg from the sites of production (soils) to the water column. A disconnect was also observed in the wetland reservoir (see St. Louis and others submitted). Water concentrations are important in the bioaccumulation of MeHg in aquatic organisms (Paterson and others 1998), so this disconnect may have important implications in MeHg contamination of reservoir fisheries.

Comparisons of FLUDEX upland and ELARP wetland reservoirs

To compare MeHg loss among upland and wetland reservoirs, mass balance budgets were calculated from June 15 to August 31 of each year. During this period of time, surface water temperatures were similar among reservoirs and above 15°C (Fig. 4-7) at which point microbial activity such as Hg methylation is enhanced (Bodaly and others 1993). In every summer post flood, the Medium C reservoir exported more MeHg per unit area (0.83 - 1.24 mg ha^{-1} day^{-1}) than the High C, Low C, and wetland reservoirs. The High C, Low C, and wetland reservoirs had very similar summer MeHg exports in the first season of flooding (Appendix 17).

Summer MeHg losses from the High C and Low C upland reservoirs (0.40 - 0.73 and 0.28 - 0.64 mg ha^{-1} day^{-1}, respectively) increased in the second year of flooding, whereas yields from the wetland reservoir (0.01 - 0.50 mg ha^{-1} day^{-1}) decreased after the first year of inundation. Summer exports of MeHg in all reservoirs were lowest in the third season of flooding. Similar trends were observed for summer THg losses. However, the export of MeHg from the reservoirs is not a good indication of overall MeHg production among the upland

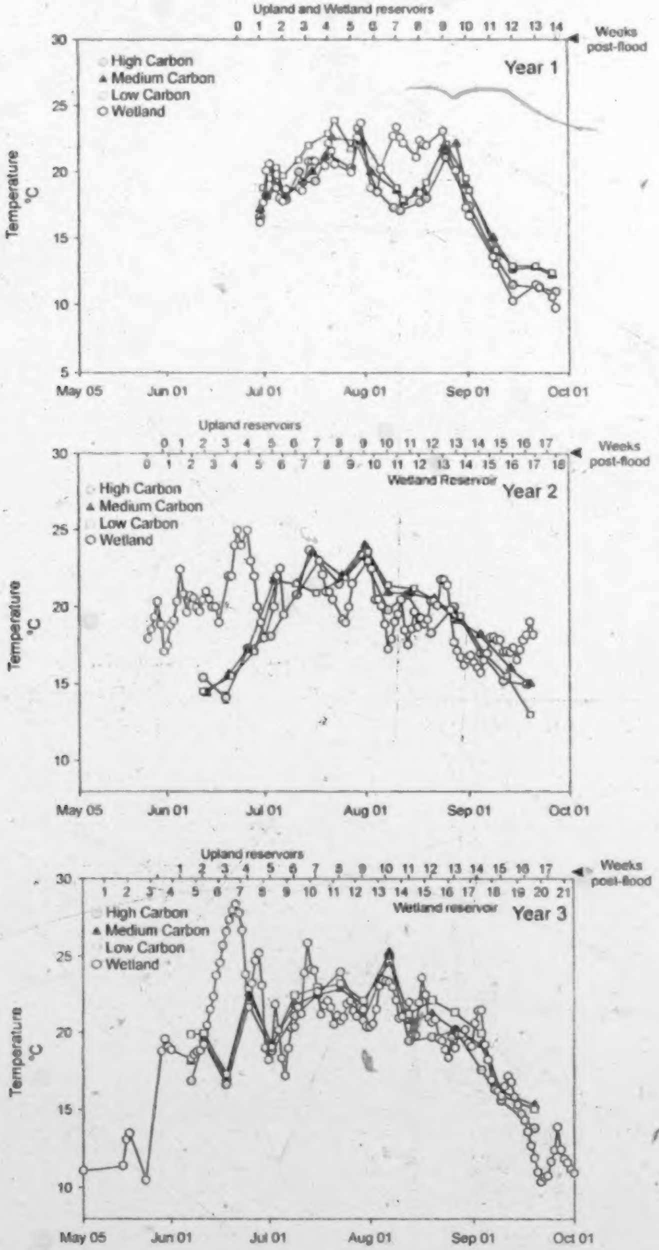

Figure 4-7. Surface water temperatures (°C) in the upland and wetland reservoirs.

and wetland reservoirs because most of the methylation occurred in the flooded soils and peat, and therefore, MeHg exported over the weir was only a small percentage of the total MeHg production. The methylation rate in the ELARP wetland reservoir was calculated by subtracting the MeHg stored in peat prior to flooding from the amount of MeHg stored in the peat two years after inundation (for details see St. Louis and others submitted).

To compare rates of methylation in the upland reservoirs with those in the wetland reservoir, we calculated similar rates of production for the first two years after flooding. Despite the ELARP wetland reservoir having 26 times more OC stores than the FLUDEX upland reservoirs, the rate of methylation in the wetland reservoir in the first two years of flooding (2700 mg ha^{-1} yr^{-1}) was only 1.7 times higher than the rate in the High C reservoir (1580 mg ha^{-1} yr^{-1}). Lower methylation rates were observed in the Medium C and Low C reservoirs at 810 and 340 mg ha^{-1} yr^{-1}, respectively. Although the highest MeHg production rate was observed in the wetland reservoir, there was more MeHg produced per unit of flooded C in the upland reservoirs (Fig. 4-8). This suggests that bacteria producing MeHg in the upland reservoirs were using OC more efficiently than microorganisms in the wetland reservoir, likely because OC stored in peat over the long term is more recalcitrant than OC stored in upland in the 17 years since the last fire.

Demethylation in the wetland reservoir was only measured in year five. The demethylation rate in the wetland (1600 mg MeHg ha^{-1} yr^{-1}) was greater than demethylation rates in the Medium C and Low C reservoirs (730 and 420 mg ha^{-1} yr^{-1}, respectively), but similar to demethylation in the High C reservoir (1440 mg ha^{-1} yr^{-1}; Fig. 4-5).

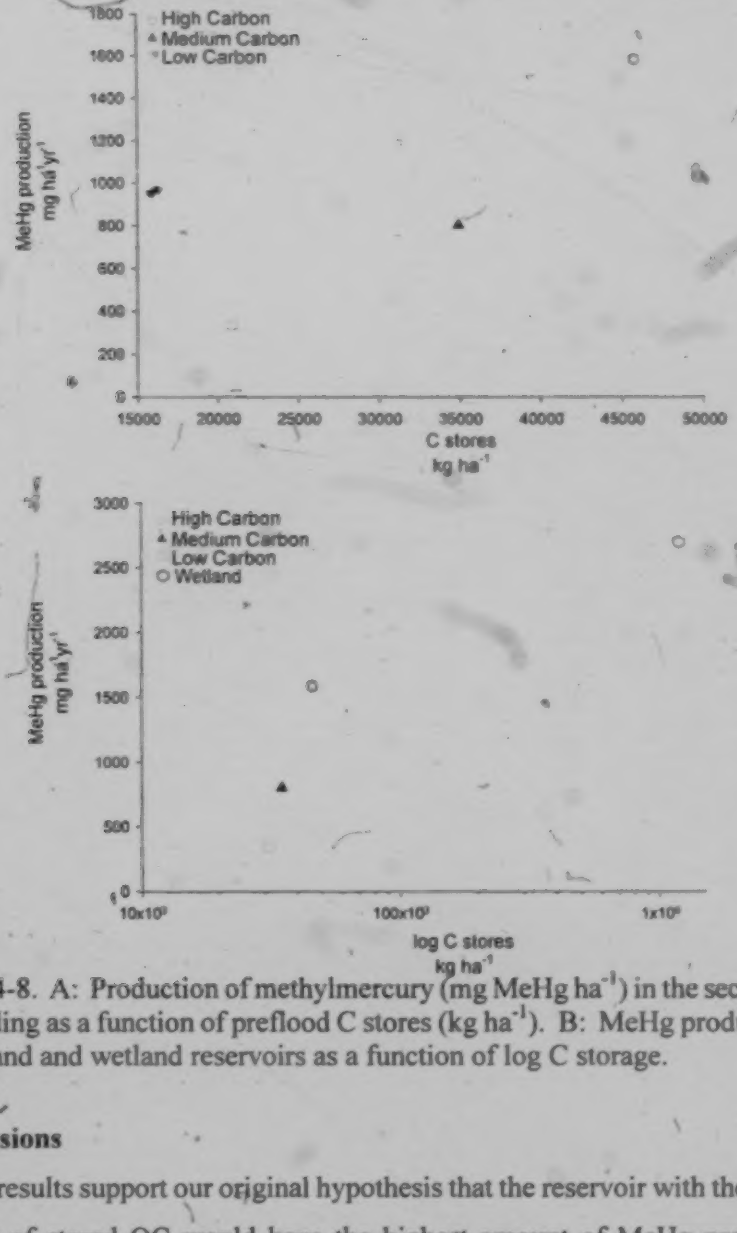

Figure 4-8. A: Production of methylmercury (mg MeHg ha^{-1}) in the second year of flooding as a function of preflood C stores (kg ha^{-1}). B: MeHg production in the upland and wetland reservoirs as a function of log C storage.

Conclusions

Our results support our original hypothesis that the reservoir with the highest *amount* of stored OC would have the highest amount of MeHg production. However, MeHg production rates in our High C reservoir were not drastically different from those in the wetland reservoir. Greenhouse gas production did not differ among the upland reservoirs (Matthews and others submitted), which indicates that the amount of easily decomposable OC was similar among the upland reservoirs. However, our results suggest that once flooded, newer, more labile OC stored in upland forests promotes higher rates of methylation compared to older, more recalcitrant OC stored in peatlands. One of the goals of the FLUDEX and ELARP reservoir projects at the ELA has been the development of computer models designed to predict MeHg increases in fish living in reservoirs. The relationship between MeHg production and total OC storage suggest that total OC stores flooded in the creation of reservoirs can be added to these models to help predict methylation rates in reservoirs.

Net MeHg production is dependant on many environmental factors (Ullrich and others 2001). Rates of production and destruction of MeHg by bacteria are affected by temperature, pH, and redox potential. However, similarities in rates of carbon GHG production in the upland reservoirs (Matthews and others submitted) suggest that the physiochemical environment did not significantly differ among the FLUDEX reservoirs, and that differences in MeHg production cannot be attributed to differences in anoxia and reducing conditions, temperature, and pH. Another important factor in the net production of MeHg is the ability of HgII to enter the cytoplasm of methylating organisms. This bioavailability could also be affected by environmental conditions, most notably the presence of inorganic and organic complexing agents (especially sulphides) that may prevent the transfer of HgII across cell membranes. Differences in SO_4 concentrations can also affect MeHg production because the majority of MeHg is produced as a by-product of metabolic SO_4 reduction. It is also possible that the bioavailability of HgII in our reservoirs differed and this resulted in differences in net MeHg production.

The majority of MeHg was produced in the soils and peat and was not transferred to the water column. Our research indicates that, unless other processes that enhance the movement of MeHg associated with flooded soils and peat particles to the water column are present (e.g. erosion see Louchouarn and others 1993), flooding wetlands does not necessarily result in a worse case scenario for MeHg contamination of reservoir fisheries because the majority of MeHg produced in the soils remains there and does not enter the water column, and thus the food web. Reservoirs created over upland forests containing relatively low OC stores may result in contamination of reservoir fisheries equal to, or exceeding, those in reservoirs created over wetland areas with very large OC stores. However, the production of MeHg and export of MeHg and THg in the upland and wetland reservoirs decreased over the first three years of flooding. This suggests that methylation rates in our reservoirs began to decrease early in the evolution of the reservoir. In fact, after only two years of flooding, there was net demethylation in the soils of the reservoirs. Regardless of declines in later years, modelling exercises have shown that 2-5 years of enhanced methylation can result in 20-30 years of elevated MeHg concentrations in predatory fish (R. Harris, Tetra Tech Inc., Oakville, ON pers. comm.). Studies on the wetland reservoir at years 4 - 9 after flooding show that MeHg concentrations in the open water region of the reservoir are not decreasing as expected (St. Louis and others submitted). Additional studies of the FLUDEX and ELARP reservoirs in 2002 and 2003 will allow us to assess further decreases in MeHg production in our experimental reservoirs.

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Chapter 5. General Summary and future research

General Summary

The FLooded Upland Dynamics EXperiment (FLUDEX) at the Experimental Lakes Area (ELA) in NW Ontario was designed to test the hypothesis that methylmercury (MeHg) and greenhouse gas production in reservoirs is related to the amount, and subsequent decomposition, of flooded organic carbon (OC) held in plants and soils. Within the framework of the FLUDEX, the overall goals of this thesis were to examine processes that contribute to increased methylation rates and to quantify, at the whole-ecosystem level, the production of MeHg in upland reservoirs.

The link between MeHg production and decomposition of flooded plant tissues was explored in detail in two studies presented in Chapters 2 and 3. Chapter 2 described an experiment in which six plastic enclosures were filled with lake water containing low concentrations of carbon (146 μmol L^{-1}) and MeHg (0.02 ng L^{-1}). The enclosures were anchored in a lake at the Experimental Lakes Area and either fresh birch leaves, fresh jack pine needles, or no plant tissues at all were added. Birch leaves decomposed 1.4 to 3.9 times faster than jack pine needles as measured by increases in dissolved inorganic carbon concentrations in enclosures. However, measured net MeHg production in enclosures containing birch leaves (0.35±0.05 ng per g carbon added) was five times lower than in the enclosures containing jack pine needles (1.94±0.28 ng per g carbon added). These results showed that MeHg production is not solely related to rates of organic matter decomposition, and that increases in MeHg associated with flooded birch leaves and jack pine needles resulted from the production of new MeHg as opposed to leaching of MeHg already in the plant tissues during decomposition.

Decomposition rates and Hg cycling in 12 different plant tissues placed in three unperturbed boreal forest sites and the three FLUDEX reservoirs over an ~800 day period were also examined using litterbags (Chapter 3). While initial rates of decomposition were slower in tissues placed in unflooded forests compared to the same tissues placed in reservoirs, there were no differences in the amount of C and N mass left in the tissues at the end of the study among sites. Depending on the initial THg concentrations in the plant tissue, decomposing plants either released THg to, took THg up from, the surrounding forests and reservoirs. In tissues that released THg, rates of decline were faster in plant tissues placed in reservoirs compared to the same plant tissue placed in unflooded forests, but there were no differences in final THg masses after 800 days. In tissues that took up THg, there were few differences between plants in forests and reservoirs. Plant tissues placed in flooded forests exhibited large increases in MeHg mass, whereas MeHg mass decreased in the same plants placed in unflooded forests. An important contribution of the litterbag study presented in Chapter 3 was to show the importance of litter decomposition to the natural biogeochemical cycling Hg in pristine forests. This was the first study to show that THg accumulates in some decomposing plant tissues in both flooded an unflooded forests and the first to measure MeHg production in decomposing boreal upland plants.

Despite the fact that MeHg production in flooded plants was not directly correlated to plant decomposition rates, the research presented in Chapter 4 supported the original hypothesis that the reservoir with the highest amount of total stored OC would have the highest amount of MeHg production. Although litterfall in upland reservoirs was not an important input of either MeHg or THg, the flux of organic carbon into the reservoirs likely stimulated increased rates of MeHg production in the litter layer of the soils. There was an initial pulse of MeHg production in all upland reservoirs in the first 2 years post flood (range =

120 - 1590 ng m^{-2} day^{-1}), followed by net demethylation (range = 360-1230 ng MeHg degraded m^{-2} day^{-1}) that reduced the pools of MeHg in the reservoirs.

Large increases in MeHg stores in soils compared to those in water and biota indicate that flooded soils were the main sites of MeHg production in the FLUDEX reservoirs. However, despite higher MeHg production in the soils in High C reservoir, MeHg concentrations in water were always higher in the Medium C reservoir compared to the other upland reservoirs and in every summer post flood, the Medium C reservoir exported more MeHg than the High C, Low C, and wetland reservoirs. This suggested that there was a disconnection between the movement of MeHg from the sites of production to the water column, and thus the aquatic food web. Unless other processes enhancing the movement of MeHg associated with flooded soils and peat particles to the water column are present, reservoirs created over upland forests containing relatively low OC stores may result in contamination of reservoir fisheries equal to, or exceeding, those in reservoirs created over wetland areas with very large OC stores.

Future research avenues

Differences in MeHg production associated with flooded OC could be due to differences in environmental factors affecting HgII bioavailability, SO_4 reduction rates, and/or rates of demethylation. For example, differences in methylation rates may be attributable to differing ability of Hg(II) to enter methylating organisms. This bioavailability could be affected by differences in environmental factors, such as concentrations of dissolved organic carbon (Barkay and others 1997), suspended particulates (such as insoluble Hg-S complexes; Benoit and others 1997), pH (Kelly and others in press), and possibly, dissolved nutrients (K. J. Scott, University of Manitoba, Winnipeg, MB pers. comm.) that can bind to Hg(II) resulting in Hg complexes that are, because of either ionic nature or size, unable to cross cell membranes. Recently, researchers have developed a genetically altered bacterium that produces light when HgII crosses the cell membrane. This tool has been used to measure the bioavailability of HgII in aquatic systems (Barkay and others 1997; Scott 2001; Golding and others 2002; Scott 2003) and would be useful in determining environmental factors that control HgII bioavailability in reservoirs.

We measured increases in the net production of MeHg in all of the enclosures and reservoirs, and therefore, methylation as opposed to demethylation was the dominant process. However, the net MeHg produced can also be affected by rates of MeHg degradation. Both reductive and oxidative biotic demethylation pathways occur in aquatic systems (Marvin-Dipasquale and others 2000). Reductive demethylation is a form of Hg resistance in bacteria possessing *mer*-operon genes, which are triggered to degrade MeHg by the presence of MeHg (Robinson and Tuovinen 1984). Oxidative demethylation is not a form of Hg resistance, but instead the co-metabolism of MeHg with the metabolism of small carbon organic substrates (Oremland and others 1991; Marvin-Dipasquale and others 2000). Both of these demethylation processes occur in anaerobic and aerobic environments by a large diversity of microbial organisms (Hobman and others 1997), however reductive demethylation results in Hg(II) and CH_4 as major end products, suggesting that it dominates in anaerobic environments, whereas the oxidative demethylation process produces CO_2 and Hg(II), and small amounts of CH_4, as end products. Future research on the production of MeHg in flooded ecosystems should include measurements of demethylation rates and the identification of the dominant demethylation process (reductive or oxidative) in reservoirs.

In the past, relative methylation and demethylation rates were determined by adding radio-isotopic ^{203}Hg or $^{14}CH_3Hg$, respectively, to sediment (Ramlal and others 1986;Gilmour and Riedel 1995) and water (Xun and others 1987) samples and measuring the by-products of methylation ($CH_3{}^{203}Hg$) and demethylation ($^{14}CO_2$ and $^{14}CH_4$). The ability to measure methylation and demethylation rates at natural concentrations was impossible because concentrations of ^{203}Hg added to samples often grossly exceeded those occurring naturally. Recently, the combination of the availability of Hg stable isotopes and ultra sensitive analytical techniques has allowed measurements of true methylation and demethylation rates at Hg concentrations that simulate natural levels (Hintelmann and others 1995). Future research on MeHg production in reservoirs should use this new technique to measure rates of methylation and demethylation on soil cores from reservoirs, allowing for the identification of specific sites of enhanced methylation or demethylation rates. For example, MeHg concentrations in soils and water were particularly high at one sampling site in the High C reservoir (K. Rolfhus, University of Wisconsin- La Crosse, La Crosse, WI pers. comm.). This site was located over a thick patch of *Sphagnum* spp. and Labrador tea and may represent a methylation 'hot spot', as describe by Branfireun and others (1996).

The experimental reservoirs created at the ELA are very different from large-scale reservoirs created in the boreal ecoregion of Canada, Scandinavia, and Russia. Large reservoirs and water diversions created to provide hydroelectricity to North America flood vast areas of land, creating deep reservoirs that have large volumes, little light attenuation, permanent stratification, and cold temperatures. The FLUDEX reservoirs were designed to mimic the shallow margins of large reservoirs, which are characterized by dead trees protruding from the water surface and experience seasonal drawdown to provide turbines with the necessary flow to meet increased energy demands in winter. The areas of flooded forest that experience drawn down may be very important methylation sites effecting Hg contamination of aquatic organisms living in open regions of the reservoir. For example, Krabbenhoft and Fink (2000) found that when inundated soils in the Florida Everglades dried out and burned, organic and inorganic sulphide compounds were oxidized to sulphate, a metabolic requirement for sulphate reducing methylating bacteria (SRB). When the soils were re-inundated, the newly liberated sulphate stimulated SRB and the efficiency of MeHg production was 10 times greater than prior to the dry down event. Therefore, MeHg production in flooded soils in upper margins of large reservoirs may contribute to long-term MeHg production and contamination of fisheries due to a fresh source of sulphate and subsequent stimulation of methylating organisms after each drawdown period. Future research should included detailed examinations of the sulphur biogeochemical cycle in reservoirs.

Finally, processes that effect the subsequent cycling of MeHg postproduction in reservoirs need to be examined in more detail. For example, the flux of MeHg from flooded soils can be affected by the flushing rate of reservoirs, erosion or the activity of benthic invertebrates and fish, and disturbance of flooded soils during seasonal drawdown.

One of the goals of the FLUDEX and ELARP reservoir experiments at the ELA has been the development of computer models designed to predict MeHg increases in fish living in reservoirs (R. Harris, Tetra Tech Inc., Oakville, ON pers. comm.). Results from my study will be used to calibrate predictive models that should assist hydroelectric utilities and government agencies in making informed decisions about selecting sites for future reservoir development to reduce MeHg contamination of the reservoir fisheries. As well, models can assist in the development of mitigation strategies that will hopefully minimize MeHg contamination in future reservoir developments.

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Appendices

Appendix 1. Concentrations of sulphate, calcium, potassium, and magnesium ions, suspended nitrogen and phosphorus, dissolved nitrogen and phosphorus, and pH, and alkalinity in unfiltered water from enclosures containing jack pine needles, birch leaves, and water only.

Appendix 2. Air-dried plant tissue mass in plant tissue in litterbags sampled from unflooded forests and reservoirs

Appendix 3. Carbon mass in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Appendix 4. Nitrogen mass in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Appendix 5. Carbon to nitrogen ratios in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Appendix 6. Total mercury mass in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Appendix 7. Methylmercury mass in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Appendix 8. Methods for measuring inputs and outputs of mercury and water.

Appendix 9. Concentrations in methylmercury and total mercury in throughfall samples taken in all three FLUDEX reservoirs (1998).

Appendix 10. Location of periphyton artificial substrates in the High C reservoir.

Appendix 11. Location of periphyton artificial substrates in the Medium C reservoir.

Appendix 12. Location of periphyton artificial substrates in the Low C reservoir.

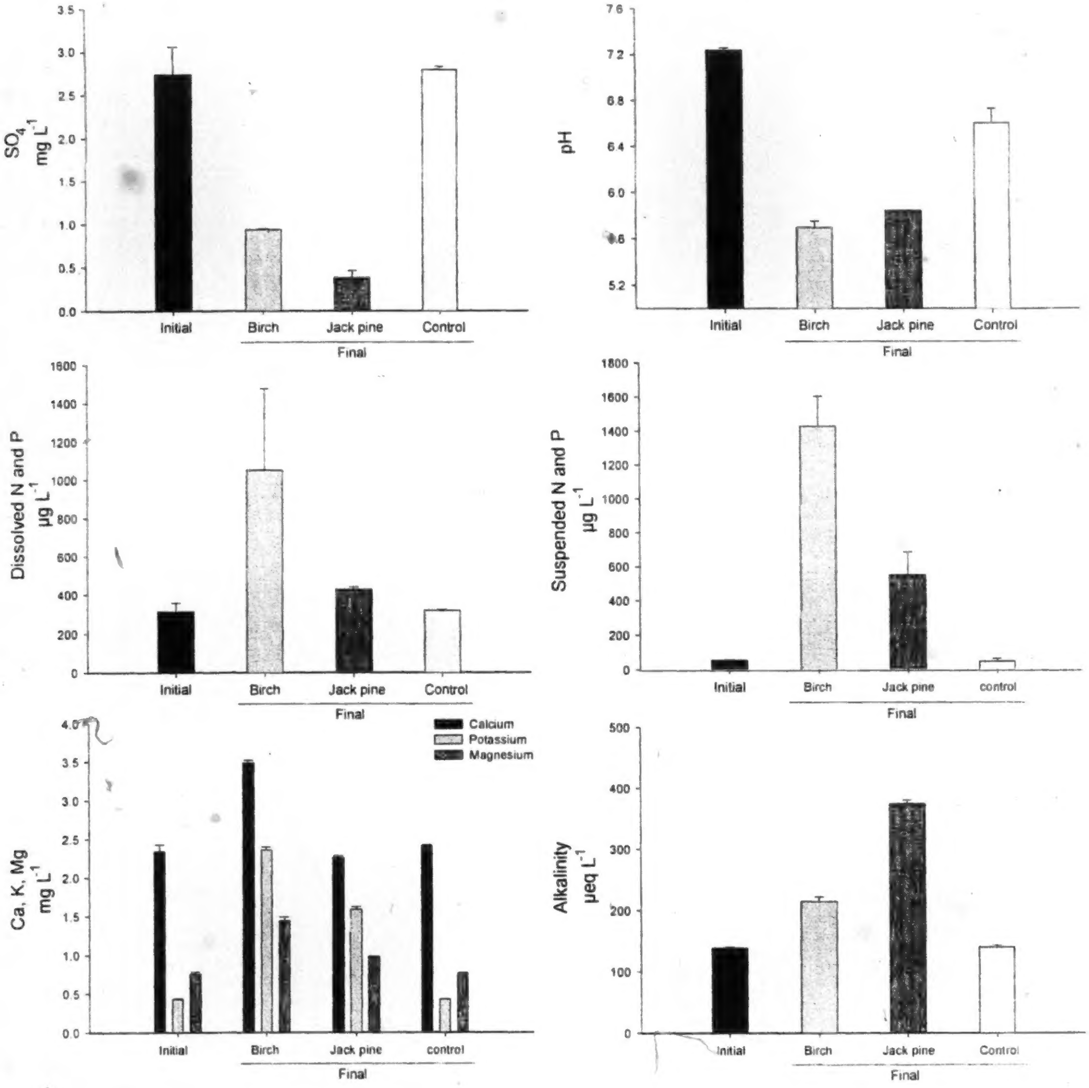
Appendix 13. Mass and carbon in litterfall falling into each FLUDEX reservoir in the first three years post flood.

Appendix 14. Concentrations of methylmercury and total mercury in wet deposition and throughfall.

Appendix 15. Average methylmercury and total mercury concentrations for wet deposition in the open and throughfall and in direct runoff.

Appendix 16. Average methylmercury and total mercury concentrations in bottom waters of upland reservoirs in September of each year.

Appendix 17. Methylmercury exports from the upland and wetland reservoirs when surface water temperatures were 15°C or higher (June 15 – August 31).



Appendix 1. Concentrations of sulphate (SO_4), calcium (Ca), potassium (K), and magnesium (Mg) ions, suspended nitrogen and phosphorus, dissolved nitrogen and phosphorus, and pH, and alkalinity in unfiltered water from enclosures containing jack pine needles, birch leaves, and water only.

Appendix 2. Air-dried plant tissue mass (g litternag^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs.

Tissue Date	High C Unflooded forest	High C Reservoir	Medium C Unflooded forest	Medium C Reservoir	Low C Unflooded forest	Low C Reservoir
Living trees						
Birch leaves						
Oct 99	2.8±0.02	2.1±0.1	3.9±0.1	1.9±0.1	4.3±0.1	1.7±0.1
May 00	2.5±0.2	2.0±0.1	3.3±0.2	1.8±0.1	3.8±0.1	1.7±0.04
Sept 00	2.1±0.1	1.7±0.2	2.2±0.2	1.7±0.1	3.5±0.02	1.4±0.1
May 01	2.1±0.1	1.9±0.1	2.0±0.1	1.7±0.1	3.4±0.1	1.4±0.04
Sept 01	1.5±0.1	2.1±0.1	1.9±0.2	1.1±0.2	3.2±0.1	1.6±0.1
Pine needles						
Oct 99	4.4±0.3	4.6±0.1	4.8±0.1	4.4±0.02	5.1±0.1	4.2±0.03
May 00	3.9±0.1	4.3±0.1	4.2±0.03	4.2±0.1	4.8±0.1	4.3±0.1
Sept 00	3.2±0.1	3.8±0.04	3.8±0.4	3.9±0.1	4.1±0.3	3.7±0.1
May 01	2.8±0.2	3.5±0.1	3.1±0.2	3.8±0.1	4.1±0.1	3.6±0.1
Sept 01	1.9±0.2	3.4±0.1	2.8±0.2	3.2±0.2	3.7±0.1	3.3±0.04
Wood						
Oct 99	13.4±0.2	14.1±1.0	12.6±0.2	12.8±0.2	12.9±0.1	12.1±0.2
May 00	12.8±0.2	12.6±0.2	13.1±0.3	12.7±0.2	12.8±0.2	12.3±0.2
Sept 00	11.9±0.3	12.2±0.2	12.4±0.5	12.5±0.4	12.5±0.2	12.1±0.2
May 01	12.8±0.8	12.6±0.2	12.7±0.1	12.4±0.2	13.0±0.6	12.4±0.1
Sept 01	11.1±0.5	12.4±0.2	10.0±0.5	12.2±0.2	12.0±0.2	12.0±0.1
Herbs and Shrubs						
Alder leaves						
Oct 99	3.3±0.1	2.7±0.1	4.1±0.1	2.2±0.1	4.3±0.1	2.2±0.03
May 00	3.0±0.1	2.6±0.1	3.3±0.3	2.2±0.04	4.0±0.1	2.1±0.1
Sept 00	2.2±0.1	2.2±0.1	2.4±0.2	2.0±0.1	2.8±0.2	1.8±0.1
May 01	2.1±0.1	2.2±0.1	2.9±0.1	2.0±0.1	3.4±0.1	2.0±0.1
Sept 01	1.8±0.1	2.3±0.1	2.4±0.03	1.6±0.1	3.0±0.2	1.7±0.04

Appendix 2 continued.

	High C		Medium C		Low C	
Tissue Date	Unflooded forest	Reservoir	Unflooded forest	Reservoir	Unflooded forest	Reservoir
Blueberry leaves						
Oct 99	3.5±0.05	2.5±0.03	3.8±0.6	2.2±0.1	3.8±0.2	2.2±0.1
May 00	3.3±0.02	2.4±0.1	3.3±0.6	2.2±0.1	3.5±0.1	2.2±0.1
Sept 00	2.4±0.02	2.1±0.1	2.7±0.1	1.9±0.1	3.2±0.1	2.0±0.1
May 01	2.6±0.05	2.2±0.1	2.6±0.4	2.0±0.02	3.1±0.02	2.0±0.1
Sept 01	2.7±0.05	2.2±0.1	2.5±0.1	1.8±0.1	2.8±0.2	1.6±0.1
Bunchberry plants						
Oct 99	2.3±0.1	1.1±0.1	3.2±0.2	0.7±0.1	nd	0.7±0.01
May 00	1.2±0.2	1.0±0.1	1.5±0.2	0.7±0.1	3.2±0.2	0.6±0.1
Sept 00	0.9±0.03	0.6±0.1	1.4±0.1	0.6±0.7	2.2±0.4	0.5±0.02
May 01	1.0±0.1	0.8±0.1	1.1±0.1	0.7±0.1	nd	0.6±0.1
Sept 01	0.9±0.1	0.8±0.03	1.0±0.1	0.4±0.1	1.2±0.003	0.4±0.02
Labrador tea leaves						
Oct 99	4.4±0.1	3.5±0.1	na	na	na	na
May 00	4.1±0.1	3.4±0.04	na	na	na	na
Sept 00	3.3±0.1	3.0±0.03	na	na	na	na
May 01	2.9±0.1	3.1±0.1	na	na	na	na
Sept 01	2.6±0.2	3.0±0.1	na	na	na	na
Bryophytes						
Sphagnum spp.						
Oct 99	4.4±0.03	4.7±0.1	na	na	na	na
May 00	4.3±0.03	4.5±0.1	na	na	na	na
Sept 00	4.3±0.1	4.5±0.03	na	na	na	na
May 01	4.7±0.2	4.6±0.1	na	na	na	na
Sept 01	3.9±0.5	4.4±0.2	na	na	na	na

Appendix 2 continued.

Tissue Date	High C Unflooded forest	High C Reservoir	Medium C Unflooded forest	Medium C Reservoir	Low C Unflooded forest	Low C Reservoir
Polytrichum spp.						
Oct 99	na	na	na	na	4.7±0.1	4.0±0.1
May 00	na	na	na	na	4.4±0.1	4.1±0.03
Sept 00	na	na	na	na	4.1±0.2	3.9±0.04
May 01	na	na	na	na	4.1±0.1	3.8±0.1
Sept 01	na	na	na	na	3.7±0.1	3.6±0.1
Pleurozium spp.						
Oct 99	na	na	na	na	4.5±0.01	4.1±0.1
May 00	na	na	na	na	4.4±0.2	4.2±0.1
Sept 00	na	na	na	na	5.1±0.5	3.9±0.03
May 01	na	na	na	na	5.5±0.4	4.2±0.1
Sept 01	na	na	na	na	4.7±0.1	3.7±0.04
Lichen						
Oct 99	na	na	na	na	5.2±0.1	3.9±0.1
May 00	na	na	na	na	4.8±0.1	3.9±0.1
Sept 00	na	na	na	na	4.5±0.2	3.2±0.3
May 01	na	na	na	na	4.9±0.1	3.0±0.3
Sept 01	na	na	na	na	5.3±0.2	2.5±0.4
'Old' wood						
Oct 99	na	na	5.1±0.1	5.3±0.1	na	na
May 00	na	na	4.5±0.6	5.4±0.3	na	na
Sept 00	na	na	5.0±0.1	4.9±0.03	na	na
May 01	na	na	5.1±0.1	5.0±0.02	na	na
Sept 01	na	na	5.0±0.1	5.0±0.03	na	na

Appendix 3. Carbon mass (g litterbag^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs. nd = do data, na= not applicable.

	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow zone	Deep zone		Shallow	Deep zone		Shallow zone	Deep zone	
Living trees									
Birch leaves									
Oct 99	1.12±0.05	1.22±0.22	1.46±0.01	1.11±0.07	1.00±0.06	2.01±0.06	1.03±0.08	0.86±0.04	2.29±0.08
May 00	0.95±0.08	1.21±0.03	1.29±0.09	0.83±0.08	1.02±0.03	1.75±0.09	0.94±0.05	0.99±0.005	1.99±0.04
Sept 00	0.75±0.09	1.04±0.05	1.03±0.04	0.89±0.12	0.87±0.03	1.14±0.09	0.76±0.05	0.72±0.05	1.78±0.01
May 01	0.94±0.11	1.01±0.03	0.88±0.12	0.82±0.03	0.83±0.08	0.97±0.03	0.67±0.03	0.73±0.01	1.41±0.30
Sept 01	nd	1.00±0.02	0.63±0.10	0.30±0.02	0.70±0.05	0.93±0.09	0.58±0.07	0.85±0.07	1.61±0.05
Pine needles									
Oct 99	2.52±0.02	2.37±0.03	2.33±0.16	2.39±0.01	2.35±0.02	2.51±0.02	2.27±0.02	2.23±0.04	2.70±0.03
May 00	2.34±0.05	2.24±0.01	2.11±0.18	3.57±1.49	2.32±0.01	2.24±0.02	2.25±0.02	2.25±0.01	2.50±0.03
Sept 00	1.92±0.04	1.98±0.05	1.65±0.05	1.98±0.03	2.05±0.05	2.05±0.21	1.79±0.08	2.12±0.06	2.09±0.12
May 01	1.80±0.04	1.49±0.41	1.48±0.09	1.93±0.06	2.05±0.71	1.58±0.15	1.87±0.05	1.73±0.18	2.15±0.07
Sept 01	nd	1.76±0.04	0.96±0.14	1.66±0.02	1.74±0.04	1.49±0.09	1.67±0.04	1.67±0.02	1.95±0.57
Wood blocks									
Oct 99	7.64±1.00	6.58±0.25	6.82±0.03	6.46±0.19	6.37±0.07	6.35±0.13	6.25±0.07	6.22±0.17	6.52±0.04
May 00	6.41±0.02	6.37±0.21	6.40±0.17	6.35±0.13	6.46±0.12	6.81±0.40	6.21±0.12	6.32±0.22	6.66±0.07
Sept 00	5.88±0.26	6.25±0.03	5.83±0.15	6.22±0.28	6.06±0.25	6.19±0.25	6.10±0.11	5.71±0.06	6.15±0.01
May 01	6.04±0.12	6.32±0.18	6.20±0.57	5.82±0.23	6.13±0.15	6.28±0.10	6.15±0.20	6.08±0.12	6.70±0.49
Sept 01	6.03±0.32	6.04±0.13	5.31±0.35	6.08±0.12	5.33±0.35	4.99±0.28	5.94±??	6.06±0.08	5.93±0.23
Herbs and Shrubs									
Alder leaves									
Oct 99	1.37±0.61	1.39±0.11	1.75±0.05	1.24±0.04	1.23±0.06	2.25±0.15	1.21±0.03	1.23±0.01	2.31±0.08
May 00	1.42±0.04	1.41±0.04	1.53±0.04	1.10±0.02	1.26±0.01	1.70±0.15	1.11±0.07	1.17±0.03	2.10±0.10
Sept 00	1.05±0.08	1.23±0.02	1.07±0.06	1.01±0.07	1.01±0.04	1.28±0.11	0.91±0.06	0.91±0.01	1.44±0.11
May 01	1.05±0.01	1.22±0.03	0.72±0.30	1.00±0.06	1.01±0.07	1.39±0.10	0.90±0.05	1.10±0.02	1.47±0.18

Appendix 3. continued.

	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow	Deep zone		Shallow zone	Deep zone		Shallow zone	Deep zone	
Sept 01	nd	1.14±0.02	0.68±0.12	0.79±0.12	0.78±0.10	1.17±0.04	0.74±0.07	0.86±0.02	1.55±0.08
Blueberry leaves									
Oct 99	1.38±0.02	1.34±0.04	1.88±0.53	1.27±0.02	1.15±0.04	2.00±0.02	1.29±0.06	1.11±0.02	2.08±0.07
May 00	1.25±0.02	1.39±0.01	1.79±0.10	0.85±0.28	1.18±0.02	1.75±0.03	1.31±0.09	1.07±0.001	1.90±0.05
Sept 00	0.98±0.03	1.16±0.01	1.23±0.11	0.96±0.07	1.04±0.04	1.42±0.07	1.04±0.03	1.01±0.04	1.32±0.04
May 01	1.04±0.02	1.23±0.04	1.33±0.06	0.97±0.02	1.02±0.03	1.34±0.20	1.05±0.02	0.93±0.10	1.60±0.05
Sept 01	nd	1.19±0.04	1.18±0.63	0.87±0.05	0.86±0.03	1.31±0.03	0.79±0.05	0.76±0.06	1.46±0.07
Bunchberry plants									
Oct 99	0.52±0.02	0.53±0.03	1.13±0.05	0.42±0.02	0.27±0.03	1.47±0.08	nd	0.35±0.01	1.47±0.05
May 00	0.46±0.04	0.57±0.02	0.58±0.08	1.25±0.93	0.37±0.05	0.68±0.11	nd	0.30±0.04	1.00±0.19
Sept 00	0.28±0.05	0.38±0.02	0.42±0.02	0.25±0.03	0.31±0.03	0.67±0.04	nd	0.26±0.01	0.56±0.003
May 01	0.30±0.01	0.41±0.01	0.46±0.05	0.32±0.02	0.33±0.05	0.51±0.04	nd	0.23±0.20	nd
Sept 01	nd	0.42±0.01	0.41±0.05	0.09±0.01	0.23±0.02	0.42±0.06	nd	0.20±0.01	nd
Labrador tea leaves									
Oct 99	2.00±0.08	1.93±0.02	2.44±0.03	na	na	na	na	na	na
May 00	1.95±0.002	1.88±0.04	2.17±0.07	na	na	na	na	na	na
Sept 00	1.59±0.002	1.62±0.03	1.58±0.14	na	na	na	na	na	na
May 01	1.65±0.07	1.65±0.06	1.45±0.03	na	na	na	na	na	na
Sept 01	nd	1.54±0.01	1.33±0.11	na	na	na	na	na	na
Bryophytes									
Sphagnum spp.									
Oct 99	2.16±0.03	2.03±0.07	2.05±0.01	na	na	na	na	na	na
May 00	2.11±0.05	1.98±0.02	1.99±0.02	na	na	na	na	na	na
Sept 00	1.85±0.02	1.80±0.01	1.85±0.02	na	na	na	na	na	na
May 01	1.95±0.01	1.87±0.06	2.03±0.012	na	na	na	na	na	na

Appendix 3. continued.

	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow	Deep zone		Shallow	Deep zone		Shallow	Deep zone	
Sept 01	0.62±0.62	1.76±0.09	1.89±0.01	na	na	na	na	na	na
Polytrichum spp.		±	±						
Oct 99	na	na	na	na	na	na	2.03±0.03	1.90±0.03	2.23±0.05
May 00	na	na	na	na	na	na	1.98±0.03	1.98±0.03	2.18±0.05
Sept 00	na	na	na	na	na	na	1.73±0.01	1.69±0.14	1.91±0.11
May 01	na	na	na	na	na	na	1.69±0.03	1.77±0.03	1.92±0.04
Sept 01	na	na	na	na	na	na	1.54±0.05	1.54±0.03	1.71±0.05
Pleurozium spp.									
Oct 99	na	na	na	na	na	na	1.97±0.01	1.90±0.06	2.37±0.04
May 00	na	na	na	na	na	na	2.06±0.11	1.94±0.02	2.14±0.03
Sept 00	na	na	na	na	na	na	1.76±0.02		2.05±0.09
May 01	na	na	na	na	na	na	1.82±0.01	1.91±0.03	2.41±0.26
Sept 01	na	na	na	na	na	na	1.64±0.05	1.61±0.02	2.45±0.24
Lichen									
Oct 99	na	na	na	na	na	na	1.80±0.09	1.83±0.12	2.28±0.16
May 00	na	na	na	na	na	na	1.94±0.20	1.76±0.06	2.19±0.04
Sept 00	na	na	na	na	na	na	1.43±0.04	1.36±0.17	1.96±0.08
May 01	na	na	na	na	na	na	1.46±0.20	1.00±0.18	1.20±0.55
Sept 01	na	na	na	na	na	na	1.41±0.15	0.67±0.04	2.05±0.24
'Old' wood									
Oct 99	na	na	na	3.23±0.03	3.03±0.06	3.01±0.03	na	na	na
May 00	na	na	na	3.00±0.46	3.01±0.03	2.68±0.37	na	na	na
Sept 00	na	na	na	2.84±0.05	2.80±0.03	2.99±0.06	na	na	na
May 01	na	na	na	2.89±0.02	2.79±0.05	2.97±0.03	na	na	na
Sept 01	na	na	na	2.83±0.01	2.85±0.06	2.84±0.08	na	na	na

Appendix 4. Nitrogen mass (g litterbag^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs. nd = do data, na= not applicable.

Tissue	High C			Medium C			Low C		
	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow	Deep zone		Shallow zone	Deep zone		Shallow zone	Deep zone	
Living trees									
Birch leaves									
Oct 99	0.10±0.01	0.10±0.004	0.10±0.002	0.10±0.01	0.06±0.01	0.12±0.003	0.09±0.010	0.06±0.003	0.13±0.004
May 00	0.09±0.01	0.12±0.004	0.09±0.01	0.07±0.01	0.08±0.001	0.12±0.004	0.06±0.03	0.07±0.003	0.11±0.009
Sept 00	0.07±0.01	0.09±0.004	0.06±0.01	0.07±0.01	0.06±0.004	0.08±0.004	0.07±0.003	0.058±0.003	0.11±0.001
May 01	0.09±0.01	0.10±0.003	0.05±0.01	0.08±0.004	0.07±0.01	0.07±0.01	0.06±0.001	0.06±0.002	0.09±0.017
Sept 01	nd	0.10±0.01	0.04±0.01	0.03±0.004	0.06±0.002	0.06±0.01	0.05±0.01	0.075±0.005	0.11±0.007
Pine needles									
Oct 99	0.07±0.004	0.065±0.002	0.06±0.010	0.07±0.001	0.06±0.001	0.06±0.001	0.07±0.003	0.06±0.002	0.066±0.001
May 00	0.07±0.01	0.06±0.001	0.07±0.001	0.07±0.0004	0.07±0.003	0.07±0.001	0.06±0.002	0.059±0.002	0.066±0.002
Sept 00	0.07±0.001	0.06±0.002	0.06±0.001	0.07±0.002	0.06±0.002	0.07±0.002	0.07±0.003	0.066±0.002	0.06±0.001
May 01	0.07±0.003	0.05±0.01	0.06±0.001	0.07±0.001	0.06±0.003	0.06±0.004	0.07±0.001	0.06±0.007	0.069±0.003
Sept 01	nd	0.06±0.001	0.05±0.01	0.07±0.001	0.06±0.001	0.06±0.003	0.06±0.003	0.058±0.0005	0.07±0.001
Wood blocks									
Oct 99	0.04±0.002	0.03±0.0004	0.04±0.007	0.03±0.0004	0.03±0.003	0.039±0.001	0.03±0.001	0.029±0.001	0.028±0.0004
May 00	0.02±0.004	0.033±0.003	0.04±0.001	0.03±0.001	0.03±0.0004	0.03±0.001	0.03±0.001	0.027±0.001	0.03±0.0001
Sept 00	0.03±0.002	0.003±0.001	0.04±0.06	0.04±0.003	0.03±0.001	0.04±0.003	0.03±0.004	0.02±0.003	0.03±0.004
May 01	0.04±0.002	0.03±0.001	0.04±0.003	0.04±0.004	0.038±0.001	0.04±0.0003	0.04±0.002	0.038±0.001	0.03±0.002
Sept 01	0.03±0.003	0.04±0.003	0.04±0.01	0.04±0.001	0.03±0.01	0.04±0.001	0.01±0.010	0.035±0.001	0.036±0.002
Herbs and Shrubs									
Alder leaves									
Oct 99	0.12±0.01	0.12±0.01	0.14±0.003	0.10±0.01	0.10±0.004	0.16±0.01	0.11±0.01	0.11±0.002	0.16±0.003
May 00	0.12±0.003	0.12±0.003	0.12±0.01	0.09±0.001	0.11±0.01	0.13±0.01	0.10±0.01	0.10±0.003	0.15±0.01
Sept 00	0.10±0.01	0.16±0.002	0.08±0.01	0.10±0.01	0.09±0.003	0.09±0.01	0.10±0.01	0.08±0.002	0.12±0.004
May 01	0.10±0.002	0.12±0.002	0.05±0.02	0.11±0.01	0.09±0.01	0.11±0.01	0.09±0.01	0.11±0.004	0.12±0.01

Appendix 4. continued.

Tissue	High C			Medium C			Low C		
	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow	Deep zone		Shallow	Deep zone		Shallow zone	Deep zone	
Blueberry leaves									
Oct 99	0.08±0.003	0.09±0.003	0.1±0.003	0.09±0.003	0.09±0.002	0.10±0.001	0.08±0.01	0.07±0.004	0.11±0.002
May 00	0.09±0.003	0.09±0.001	0.10±0.003	0.08±0.01	0.08±0.002	0.10±0.002	0.09±0.01	0.07±0.000	0.10±0.004
Sept 00	0.08±0.07	0.08±0.003	0.08±0.01	0.08±0.01	0.08±0.001	0.09±0.01	0.08±0.004	0.07±0.01	0.10±0.001
May 01	0.08±0.002	0.09±0.01	0.08±0.01	0.08±0.002	0.08±0.001	0.09±0.01	0.08±0.004	0.07±0.004	0.10±0.001
Sept 01	nd	0.10±0.01	0.08±0.001	0.07±0.01	0.07±0.002	0.08±0.01	0.07±0.01	0.058±0.00	0.09±0.002
Bunchberry plants									
Oct 99	0.06±0.004	0.05±0.003	0.10±0.004	0.04±0.001	0.02±0.004	0.11±0.01	nd	0.03±0.002	nd
May 00	0.04±0.01	0.06±0.002	0.04±0.01	0.02±0.003	0.03±0.001	0.06±0.01	nd	0.02±0.003	0.1±0.003
Sept 00	0.02±0.01	0.03±0.002	0.03±0.001	0.02±0.002	0.02±0.004	0.05±0.004	nd	0.02±0.001	0.07±0.0
May 01	0.03±0.001	0.04±0.003	0.04±0.01	0.03±0.002	0.02±0.01	0.04±0.003	nd	0.02±0.002	nd
Sept 01	nd	0.04±0.002	0.03±0.004	0.01±0.001	0.02±0.002	0.04±0.004	nd	0.02±0.001	0.05±0.002
Labrador tea leaves									
Oct 99	0.08±0.01	0.08±0.001	0.08±0.003	na	na	na	na	na	na
May 00	0.08±0.003	0.07±0.003	0.08±0.0003	na	na	na	na	na	na
Sept 00	0.07±0.001	0.06±0.002	0.07±0.01	na	na	na	na	na	na
May 01	0.08±0.01	0.09±0.001	0.07±0.002	na	na	na	na	na	na
Sept 01	nd	0.07±0.003	0.06±0.004	na	na	na	na	na	na
Bryophytes									
Sphagnum spp.									
Oct 99	0.05±0.003	0.03±0.002	0.05±0.003	na	na	na	na	na	na
May 00	0.03±0.001	0.03±0.01	0.04±0.002	na	na	na	na	na	na
Sept 00	0.04±0.001	0.03±0.002	0.03±0.004	na	na	na	na	na	na
May 01	0.04±0.001	0.04±0.002	0.05±0.002	na	na	na	na	na	na

Appendix 4. continued.

	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow zone	Deep zone		Shallow zone	Deep zone		Shallow	Deep zone	
Sept 01	0.01±0.01	0.03±0.01	0.04±0.001	na	na	na	na	na	na
Polytrichum spp.									
Oct 99	na	na	na	na	na	na	0.06±0.002	0.046±0.002	0.08±0.002
May 00	na	na	na	na	na	na	0.05±0.003	0.045±0.004	0.074±0.001
Sept 00	na	na	na	na	na	na	0.05±0.002	0.038±0.004	0.064±0.009
May 01	na	na	na	na	na	na	0.05±0.001	0.04±0.001	0.07±0.004
Sept 01	na	na	na	na	na	na	0.04±0.004	0.036±0.0003	0.059±0.004
Pleurozium spp.									
Oct 99	na	na	na	na	na	na	0.04±0.002	0.03±0.001	0.05±0.004
May 00	na	na	na	na	na	na	0.04±0.002	0.03±0.001	0.055±0.002
Sept 00	na	na	na	na	na	na	0.03±0.002	0.036	0.05±0.003
May 01	na	na	na	na	na	na	0.04±0.001	0.035±0.001	0.065±0.005
Sept 01	na	na	na	na	na	na	0.03±0.002	0.02±0.0004	0.059±0.003
Lichen									
Oct 99	na	na	na	na	na	na	0.03±0.002	0.02±0.003	0.029±0.002
May 00	na	na	na	na	na	na	0.1±0.05	0.019±0.001	0.026±0.002
Sept 00	na	na	na	na	na	na	0.02±0.004	0.019±0.002	0.02±0.001
May 01	na	na	na	na	na	na	0.02±0.002	0.017±0.003	0.018±0.008
Sept 01	na	na	na	na	na	na	0.02±0.002	0.01±0.001	0.035±0.010
'Old' wood									
Oct 99	na	na	na	0.02±0.001	0.02±0.002	0.02±0.001	na	na	na
May 00	na	na	na	0.02±0.0	0.02±0.001	0.01±0.002	na	na	na
Sept 00	na	na	na	0.02±0.001	0.02±0.001	0.02±0.001	na	na	na
May 01	na	na	na	0.02±0.001	0.02±0.001	0.02±0.001	na	na	na
Sept 01	na	na	na	0.02±0.0	0.02±0.001	0.02±0.004	na	na	na

Appendix 5. Carbon to nitrogen ratios in plant tissue in litterbags sampled from unflooded forests and reservoirs. nd = do data, na= not applicable.

	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow zone	Deep zone		Shallow zone	Deep zone		Shallow zone	Deep zone	
Living trees									
Birch leaves									
Oct 99	12.01±1.11	11.93±0.29	13.97±0.28	11.57±0.17	15.89±0.29	17.16±0.94	12.29±0.90	14.08±0.78	17.45±1.30
May 00	11.07±0.41	10.15±0.56	14.36±0.37	11.69±0.55	12.44±0.18	14.74±0.27	10.70±0.76	13.79±0.44	17.98±0.04
Sept 00	11.24±0.41	11.07±0.12	17.28±1.36	13.27±0.45	14.28±0.94	15.04±0.47	10.97±0.48	12.27±0.47	15.58±0.62
May 01	11.09±0.49	10.22±0.14	16.75±0.69	10.03±0.16	11.40±0.49	12.23±0.88	11.47±0.52	11.91±0.41	15.30±0.56
Sept 01	nd	10.41±0.32	15.46±0.07	11.53±1.00	11.86±0.36	16.05±0.45	11.81±0.18	11.24±0.27	14.51±0.48
Pine needles									
Oct 99	34.92±1.51	36.63±0.68	38.63±4.39	36.20±0.46	37.31±0.84	39.06±0.88	34.80±1.99	35.38±0.51	40.91±0.16
May 00	33.23±2.22	37.51±0.66	29.95±0.48	31.11±0.49	35.91±1.81	34.70±0.34	37.43±1.53	38.18±1.30	38.02±1.19
Sept 00	28.11±0.56	31.76±0.65	25.75±1.25	29.18±0.67	32.77±0.31	28.00±3.53	26.72±0.71	31.83±0.25	34.20±1.92
May 01	26.60±0.66	27.50±1.64	23.07±1.02	26.70±0.55	32.88±2.59	26.44±2.06	26.95±0.86	28.69±0.24	31.20±0.78
Sept 01	nd	29.93±1.04	20.44±1.06	23.34±0.26	28.40±0.98	25.15±0.33	26.43±0.83	29.02±0.48	27.81±0.29
Wood blocks									
Oct 99	212.58±13.84	218.31±6.42	169.44±24.32	236.45±2.80	218.82±19.88	218.06±13.48	230.14±6.54	218.50±17.27	233.36±3.84
May 00	462.29±135.56	197.88±16.84	179.27±8.89	223.04±4.82	218.96±0.84	220.00±11.33	216.32±2.67	233.23±3.36	216.56±5.59
Sept 00	182.18±15.70	253.07±9.26	142.36±17.42	174.46±9.17	201.76±10.97	164.94±17.87	189.08±16.53	243.77±8.88	208.55±25.20
May 01	156.23±9.02	180.51±19.55	143.15±4.56	163.68±11.41	159.42±3.50	177.20±2.02	153.73±12.88	160.63±10.31	209.17±9.47
Sept 01	177.42±14.43	167.77±7.36	140.11±27.56	164.45±5.68	166.24±17.66	144.74±8.32	205.67	175.19±6.12	161.66±12.73
Herbs and Shrubs									
Alder leaves									
Oct 99	11.84±0.06	11.40±0.40	12.89±0.16	12.32±0.82	11.86±0.18	13.96±0.42	11.64±0.55	11.55±0.37	14.78±0.18
May 00	11.59±0.04	11.47±0.61	13.14±0.18	11.92±0.07	11.66±0.46	12.81±0.09	11.72±0.61	12.11±0.20	13.92±0.31
Sept 00	10.92±0.24	10.67±0.06	14.28±0.63	10.24±0.29	11.25±0.23	13.90±0.74	9.54±0.17	11.47±0.11	12.31±0.48
May 01	10.29±0.10	10.01±0.05	13.46±0.57	9.62±0.27	11.06±0.16	11.70±0.08	10.06±0.65	10.29±0.34	12.58±0.46

Appendix 5. continued.

Tissue	High C			Medium C			Low C		
	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow	Deep zone		Shallow	Deep zone		Shallow	Deep zone	
Sept 01	nd	11.50±1.44	16.36±0.98	10.09±0.16	10.39±0.41	14.98±1.88	9.55±0.11	10.00±0.24	12.31±1.02
Blueberry leaves									
Oct 99	14.23±0.21	15.02±0.05	17.54±0.06	14.46±0.24	13.03±0.16	19.85±0.42	16.28±0.46	16.18±0.63	19.00±0.48
May 00	14.53±0.31	14.77±0.12	17.82±0.63	13.79±1.68	15.40±0.58	17.49±0.49	13.65±0.83	15.32±0.03	18.45±0.35
Sept 00	12.85±0.46	15.48±0.68	16.43±0.63	11.66±0.56	13.59±0.45	15.42±0.14	13.41±0.44	14.14±0.45	17.21±0.28
May 01	13.16±0.13	13.18±0.44	16.82±0.33	11.75±0.18	13.59±0.26	14.79±1.11	12.67±0.53	13.33±0.03	16.19±0.32
Sept 01	nd	12.46±1.13	15.63±0.16	12.81±0.20	12.92±0.09	15.78±0.67	11.93±0.09	12.99±0.38	16.19±0.31
Bunchberry plants									
Oct 99	9.51±0.39	11.21±0.17	11.49±0.15	11.03±0.21	11.93±1.35	13.40±0.24	nd	12.59±1.12	nd
May 00	11.82±0.68	9.78±0.40	13.66±0.26	13.15±0.22	13.43±0.85	11.86±0.20	nd	16.92±1.02	13.54±0.26
Sept 00	12.46±1.11	12.58±0.43	13.41±0.25	12.43±0.22	15.40±1.67	12.57±0.28	nd	15.20±0.63	13.33±0.99
May 01	11.23±0.59	11.51±0.57	12.89±0.50	10.99±0.27	14.22±1.46	4.53±0.13	nd	15.05±0.98	nd
Sept 01	nd	11.96±0.40	13.10±0.58	14.91±1.85	14.34±0.74	12.89±0.40	nd	12.91±0.17	12.34±0.58
Labrador tea leaves									
Oct 99	24.65±0.88	25.60±0.39	45.70±2.46	na	na	na	na	na	na
May 00	24.84±0.93	26.80±0.90	49.80±1.75	na	na	na	na	na	na
Sept 00	21.59±0.38	25.44±0.35	57.88±6.11	na	na	na	na	na	na
May 01	21.06±1.99	19.21±0.48	42.21±0.22	na	na	na	na	na	na
Sept 01	nd	22.25±2.09	43.74±0.74	na	na	na	na	na	na
Bryophytes									
Sphagnum spp.									
Oct 99	47.11±3.96	60.82±3.38	28.99±0.60	na	na	na	na	na	na
May 00	80.32±0.34	52.94±1.76	25.93±0.89	na	na	na	na	na	na
Sept 00	49.52±1.65	53.03±3.74	23.09±0.11	na	na	na	na	na	na
May 01	49.15±1.78	53.40±1.97	21.83±0.51	na	na	na	na	na	na

Appendix 5. continued.

	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow zone	Deep zone		Shallow zone	Deep zone		Shallow zone	Deep zone	
Sept 01	nd	57.23±10.73	23.07±0.83	na	na	na	na	na	na
Polytrichum spp.									
Oct 99	na	na	na	na	na	na	34.89±1.65	41.39±1.73	27.71±0.17
May 00	na	na	na	na	na	na	38.29±1.51	44.34±2.82	29.43±0.09
Sept 00	na	na	na	na	na	na	35.22±1.34	45.29±1.19	30.08±2.70
May 01	na	na	na	na	na	na	36.74±0.46	41.12±1.32	27.49±0.99
Sept 01	na	na	na	na	na	na	35.80±2.01	42.19±0.69	29.40±2.50
Pleurozium spp.									
Oct 99	na	na	na	na	na	na	55.29±3.70	64.72±3.63	44.32±3.91
May 00	na	na	na	na	na	na	55.33±0.18	65.23±2.33	39.13±1.01
Sept 00	na	na	na	na	na	na	52.91±3.19	62.41	39.28±0.97
May 01	na	na	na	na	na	na	52.52±1.91	55.36±1.16	37.17±2.55
Sept 01	na	na	na	na	na	na	49.99±3.85	74.56±2.32	41.17±2.03
Lichen									
Oct 99	na	na	na	na	na	na	73.07±1.35	83.68±13.23	79.65±2.43
May 00	na	na	na	na	na	na	48.43±18.67	90.59±2.28	83.93±7.04
Sept 00	na	na	na	na	na	na	72.94±7.70	71.94±6.84	87.14±5.13
May 01	na	na	na	na	na	na	61.43±4.32	59.56±2.82	65.95±1.37
Sept 01	na	na	na	na	na	na	62.91±7.57	61.65±7.40	64.03±9.94
'Old' wood									
Oct 99	na	na	na	207.81±8.55	191.63±5.75	189.84±12.49	na	na	na
May 00	na	na	na	152.95±24.82	200.63±8.27	192.96±3.34	na	na	na
Sept 00	na	na	na	180.48±11.98	184.37±4.42	170.16±8.60	na	na	na
May 01	na	na	na	154.93±0.76	158.88±11.00	161.55±10.22	na	na	na
Sept 01	na	na	na	150.98±3.71	167.61±6.82	153.84±5.16	na	na	na

Appendix 6. Total mercury mass (ng THg litterbag^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs.in litterbags. nd = do data, na= not applicable.

Tissue	High C			Medium C			Low C		
	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow zone	Deep zone		Shallow zone	Deep zone		Shallow zone	Deep zone	
Living trees									
Birch leaves									
Oct 99	138.43±6.67	61.76±1.45	49.33±0.30	81.11±4.89	46.03±2.79	45.27±1.32	68.80±5.61	53.77±1.77	45.47±1.45
Sept 00	99.96±11.35	77.11±3.72	72.42±2.58	111.49±14.50	59.78±1.43	91.81±7.53	66.30±3.64	59.43±3.45	53.48±0.25
Sept 01	88.93±22.50	124.96±3.60	72.93±6.07	69.24±4.71	77.03±4.27	111.05±10.5	88.65±3.22	66.69±5.43	51.43±1.95
Pine needles									
Oct 99	82.65±0.38	57.32±0.52	69.29±4.69	66.65±0.23	59.49±0.38	65.62±0.73	61.61±0.25	71.15±1.16	63.34±1.67
Sept 00	71.49±1.07	65.89±0.38	84.16±3.14	80.00±1.43	66.80±1.59	85.25±9.85	68.57±1.41	58.71±0.27	67.98±4.29
Sept 01	85.42±0.82	95.32±1.63	82.17±9.16	88.71±1.51	78.70±0.63	111.54±6.55	83.53±1.68	72.89±0.90	87.76±2.59
Wood blocks									
Oct 99	62.40±8.21	36.74±0.83	43.78±0.60	50.00±1.58	56.53±0.69	33.54±0.47	28.83±0.10	39.51±0.93	40.02±0.29
Sept 00	79.19±1.02	77.48±1.77	145.20±3.18	59.18±1.84	78.77±1.48	57.02±2.20	89.07±1.75	55.52±0.91	66.90±1.15
Sept 01	65.53±2.05	68.51±0.74	130.17±6.23	56.80±1.38	54.51±1.07	77.15±3.74	45.20±0.69	51.01±0.57	69.80±1.37
Herbs and Shrubs									
Alder leaves									
Oct 99	131.58±0.34	55.86±1.49	48.23±1.18	69.92±2.30	60.52±3.20	59.31±1.91	60.73±1.87	58.81±0.73	42.93±1.06
Sept 00	104.80±6.38	81.54±0.75	74.24±3.49	79.21±4.84	81.32±2.11	82.53±7.13	74.69±4.27	68.83±0.81	41.52±3.04
Sept 01	115.89±1.40	110.69±3.31	53.11±3.83	96.21±13.10	72.25±5.55	97.83±1.42	78.55±6.53	84.37±1.76	91.06±4.83
Blueberry leaves									
Oct 99	122.33±2.26	59.16±1.18	37.68±1.54	57.72±1.54	47.71±1.68	42.35±0.64	64.28±2.36	57.78±0.99	34.98±1.41
Sept 00	96.60±6.56	88.64±0.52	56.69±4.94	103.68±5.64	73.89±1.24	70.82±3.11	66.22±1.41	72.36±2.90	69.85±1.05
Sept 01	136.60±2.44	112.50±3.16	78.24±1.77	110.91±7.13	77.72±1.26	101.19±1.87	91.851±6.41	57.52±3.36	73.54±3.87
Bunchberry plants									
Oct 99	137.51±17.16	60.14±1.52	44.23±0.63	70.49±3.40	35.25±3.38	37.10±2.54	nd	58.77±1.07	nd
Sept 00	41.21±6.21	34.94±1.61	35.05±1.23	39.51±4.95	47.77±5.56	57.56±3.19	nd	35.22±1.19	61.97±12.33
Sept 01	37.90±2.31	41.52±1.43	57.17±1.29	32.82±5.13	31.97±1.10	54.85±2.51	nd	28.27±1.35	54.53±0.16

Appendix 6. continued.

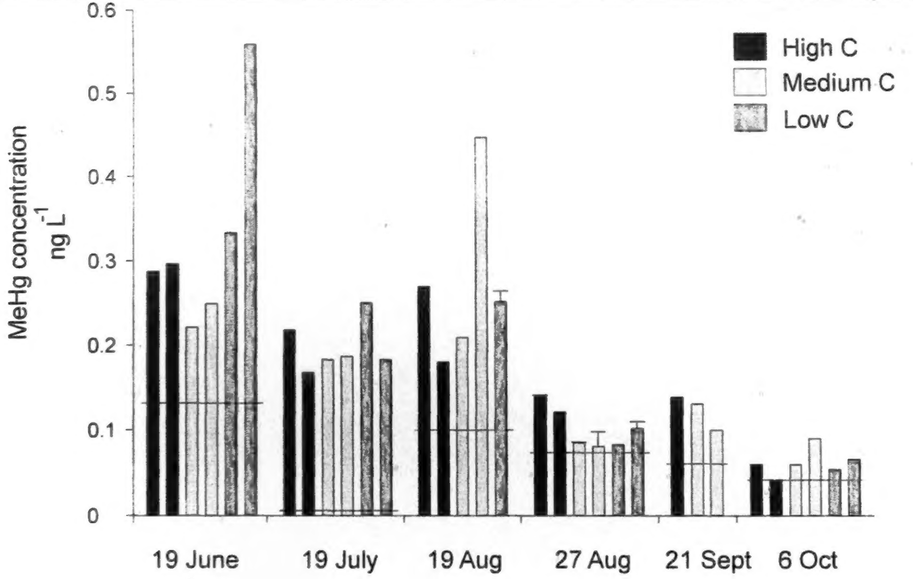
	High C			Medium C			Low C		
Tissue	Reservoir		Forested Reference	Reservoir		Forested Reference	Reservoir		Forested Reference
Date	Shallow zone	Deep zone		Shallow zone	Deep zone		Shallow zone	Deep zone	
Labrador tea leaves									
Oct 99	210.17±2.81	103.83±1.31	249.91±1.90	na	na	na	na	na	na
Sept 00	150.09±1.91	121.13±1.34	164.71±2.79	na	na	na	na	na	na
Sept 01	103.90±2.43	134.92±6.17	141.08±9.11	na	na	na	na	na	na
Bryophytes									
Sphagnum spp.									
Oct 99	176.22±7.54	113.18±0.50	50.42±2.04	na	na	na	na	na	na
Sept 00	121.73±0.73	124.86±0.73	98.53±2.38	na	na	na	na	na	na
Sept 01	182.15±6.22	155.06±2.95	170.05±12.33	na	na	na	na	na	na
Polytrichum spp.									
Oct 99	na	na	na	na	na	na	162.47±1.02	178.98±2.50	235.51±4.57
Sept 00	na	na	na	na	na	na	166.56±3.33	130.79±0.97	251.06±14.85
Sept 01	na	na	na	na	na	na	189.89±7.43	138.37±2.11	275.60±8.22
Pleurozium spp.									
Oct 99	na	na	na	na	na	na	168.90±0.98	143.47±3.06	288.97±4.05
Sept 00	na	na	na	na	na	na	178.23±1.40	128.89	292.04±12.95
Sept 01	na	na	na	na	na	na	206.72±3.90	129.50±1.70	295.38±18.96
Lichen									
Oct 99	na	na	na	na	na	na	121.57±7.79	110.61±1.56	136.59±2.74
Sept 00	na	na	na	na	na	na	74.12±1.88	71.33±12.35	131.52±5.52
Sept 01	na	na	na	na	na	na	59.98±7.01	48.46±2.62	131.02±4.22
'Old' wood									
Oct 99	na	na	na	45.19±0.54	52.57±0.96	35.37±0.34	na	na	na
Sept 00	na	na	na	49.89±0.60	73.65±0.23	39.37±0.74	na	na	na
Sept 01	na	na	na	76.72±0.36	74.36±0.38	46.29±1.06	na	na	na

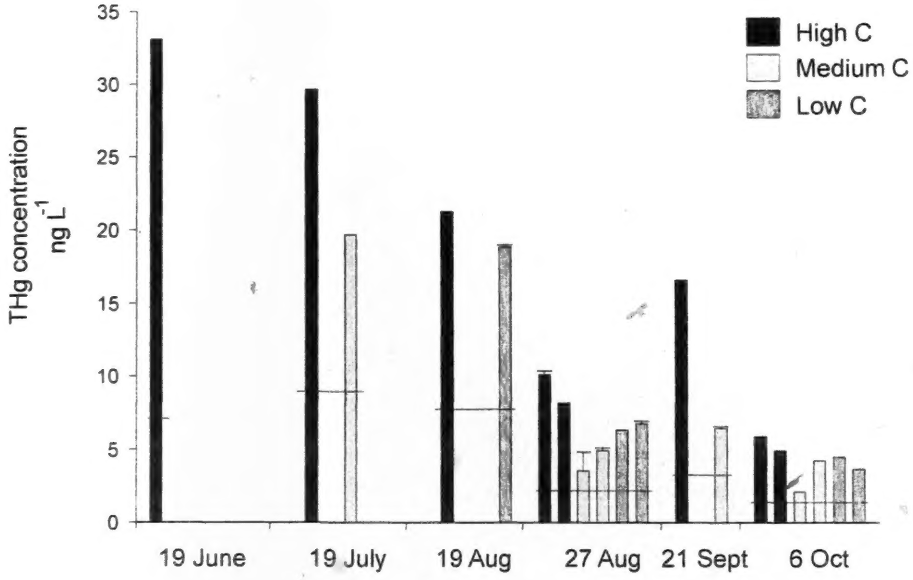
Appendix 7. Methylmercury mass (ng MeHg litterbag^{-1}) in plant tissue in litterbags sampled from unflooded forests and reservoirs. nd = do data, na= not applicable.

	High C		Medium C		Low C	
Tissue Date	Reservoir (Deep zone)	Forested Reference	Reservoir (Deep zone)	Forested Reference	Reservoir (Deep zone)	Forested Reference
Living trees						
Pine needles						
Oct 99	3.22±0.03	0.35±0.02	1.19±0.01	0.19±0.00	1.48±0.02	0.39±0.01
Sept 00	2.59±0.01	undetectable	3.25±0.08	0.50±0.06	2.01±0.01	0.29±0.02
Sept 01	3.79±0.07	1.79±0.20	5.41±0.04	0.77±0.05	2.52±0.03	1.04±0.03
Herbs and Shrubs						
Blueberry leaves						
Oct 99	12.74±0.25	3.00±0.04	3.10±0.11	0.51±0.01	6.89±0.12	0.43±0.02
Sept 00	9.87±0.06	0.38±0.03	7.59±0.13	0.60±0.03	4.15±0.17	0.74±0.01
Sept 01	18.99±0.53	1.66±0.04	12.45±0.20	0.57±0.01	8.14±0.47	0.71±0.04
Labrador tea leaves						
Oct 99	4.70±0.02	1.91±0.03	na	na	na	na
Sept 00	5.87±0.03	1.77±0.04	na	na	na	na
Sept 01	18.69±0.36	0.72±0.05	na	na	na	na
Bryophytes						
Polytrichum						
Oct 99	na	na	na	na	7.72±0.11	0.68±0.01
Sept 00	na	na	na	na	15.73±0.1	1.93±0.11
Sept 01	na	na	na	na	19.96±0.3	1.46±0.04
Pleurozium						
Oct 99	na	na	na	na	22.33±0.4	9.25±0.13
Sept 00	na	na	na	na	8.92	2.66±0.12
Sept 01	na	na	na	na	16.85±0.2	5.02±0.32
Lichen						
Oct 99	na	na	na	na	3.74±0.05	1.37±0.03
Sept 00	na	na	na	na	2.42±0.30	0.44±0.02
Sept 01	na	na	na	na	2.50±0.14	1.91±0.06

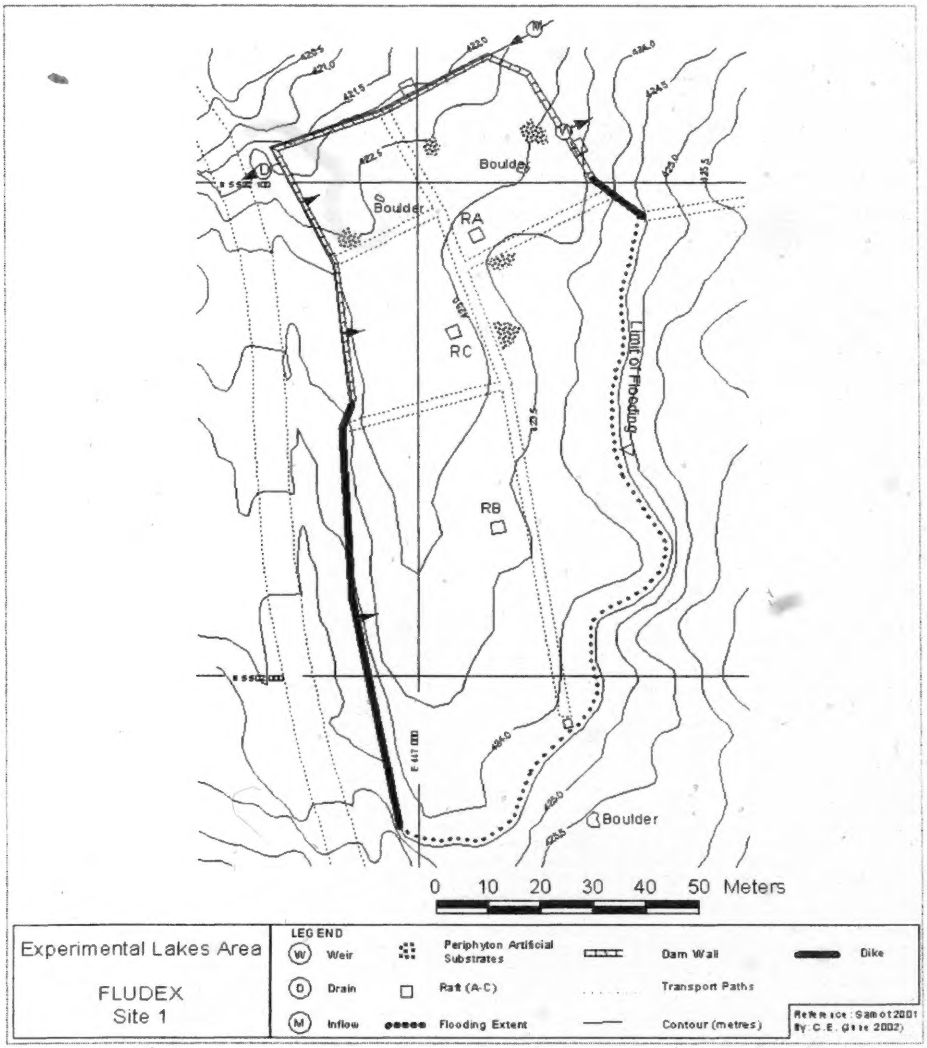
Appendix 8. Methods for measuring inputs and outputs of mercury and water.

Parameter	Method of measurement
Inputs	
Pumped water from Roddy Lake	Weekly and biweekly concentrations measured at point of water entry to reservoirs; Daily water volumes measured by inline flow meters
Throughfall	Concentrations measured in throughfall in reservoirs; Water volume calculated by multiplying the total precipitation by a canopy interception factor measured in the reservoirs
Wet deposition in the open	Concentrations measured in wet deposition collected at the ELA meteorological site and the Lake 658 watershed; Daily water volumes measured at the ELA meteorological site
Direct runoff	Concentrations measured in runoff from a purely upland catchment (Lake 114 upland); Water volumes estimated on an aerial basis from a gauged watershed (NW inflow to Rawson Lake) near the upland reservoirs
Litterfall	Weight of foliage in canopy were multiplied by concentrations of MeHg and THg in jack pine forest litter from a previous study at the ELA
Outputs	
Outflow over weir	Weekly and biweekly concentrations measured at the outflow of each reservoir; Water volumes measured daily at weir
Seepage	Concentrations measured at weir outflow; Volume of seepage from each reservoir was taken as the residual volume and verified by independent seepage measurement surveys performed periodically during the study period.
Drawdown pipe	
Surface water	Concentrations measured at weir; Volumes of surface waters were calculated as 95% of total volume of each reservoir (estimated based on aerial mapping)
Bottom water	Concentrations measured in bottom waters; Volumes of bottom waters were calculated as 5% of total volume of each reservoir (estimated based on aerial mapping)
Hg^0 evasion	Dissolved gaseous elemental Hg was measured by purging 2L of reservoir water with ultra high purity nitrogen onto gold-bead Hg traps

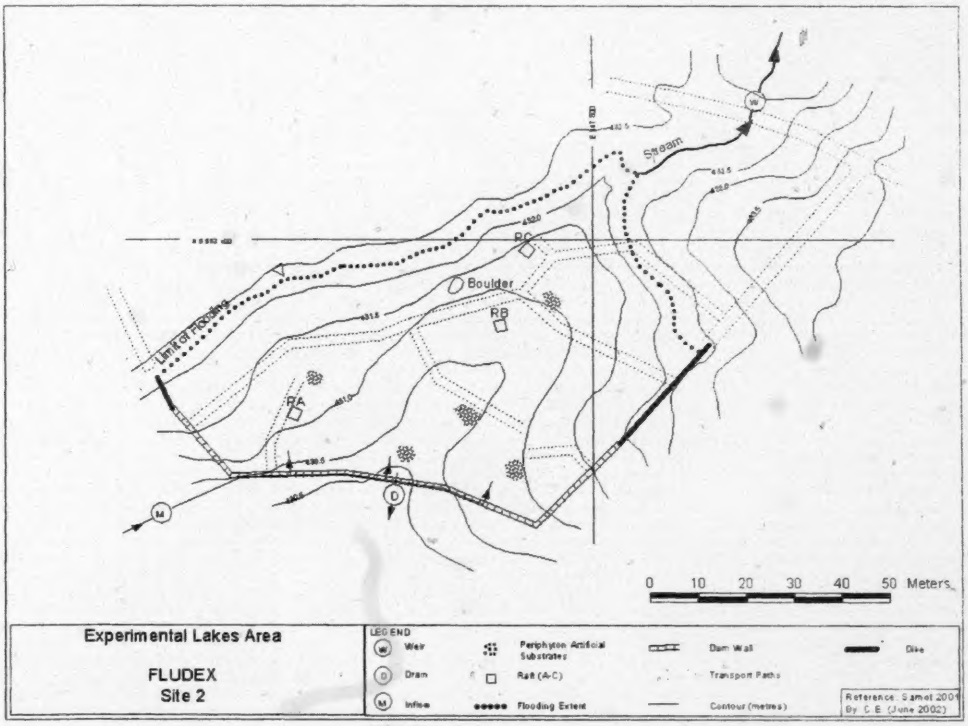




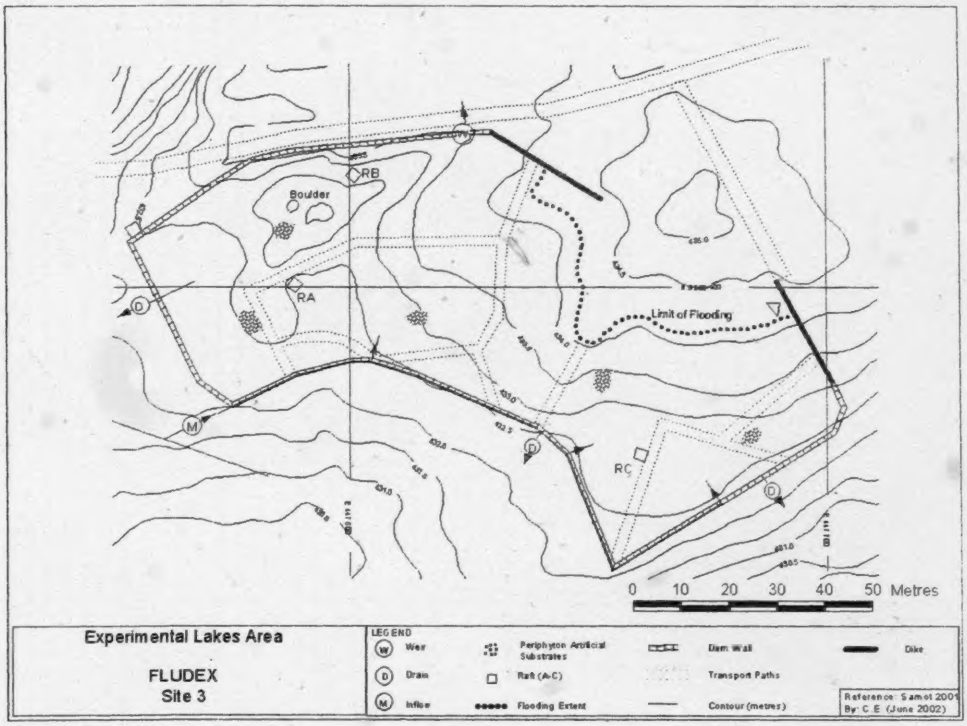
Appendix 9. Methylmercury (MeHg) and total mercury (THg) concentrations (ng L^{-1}) in throughfall samples taken in all three FLUDEX reservoirs (1998). Horizontal lines represent concentrations in precipitation in the open areas.


M
W
D
Boulder
Boulder
Boulder
RA
RC
RB
Limit of Flooding
424.0
424.5
425.0
425.5
421.0
421.5
422.0
422.5
423.0
423.5
E 447 000
0 10 20 30 40 50 Meters
Experimental Lakes Area
FLUDEX
Site 1
LEGEND
W Weir
D Drain
M Inflow
Periphyton Artificial Substrates
Raft (A-C)
Flooding Extent
Dam Wall
Transport Paths
Contour (metres)
Dike
Reference : Samot 2001

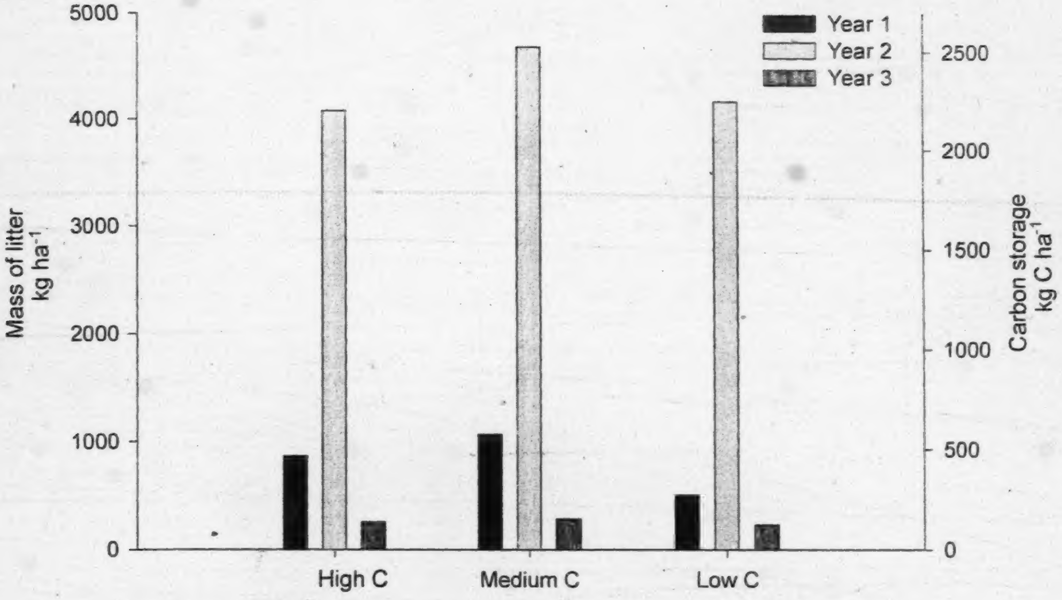
Appendix 10. Locations of artificial substrates for sampling periphyton from the High C reservoir.


Stream
Boulder
Limit of Flooding
RA
RB
RC
W
D
M
432.5
432.0
431.5
431.0
430.5
0 10 20 30 40 50 Meters
Experimental Lakes Area
FLUDEX
Site 2
LEGEND
Weir
Drain
Inflow
Periphyton Artificial Substrates
Raft (A-C)
Flooding Extent
Dam Wall
Transport Paths
Contour (metres)
Dike
Reference: Samot 2001
By: C.E. (June 2002)

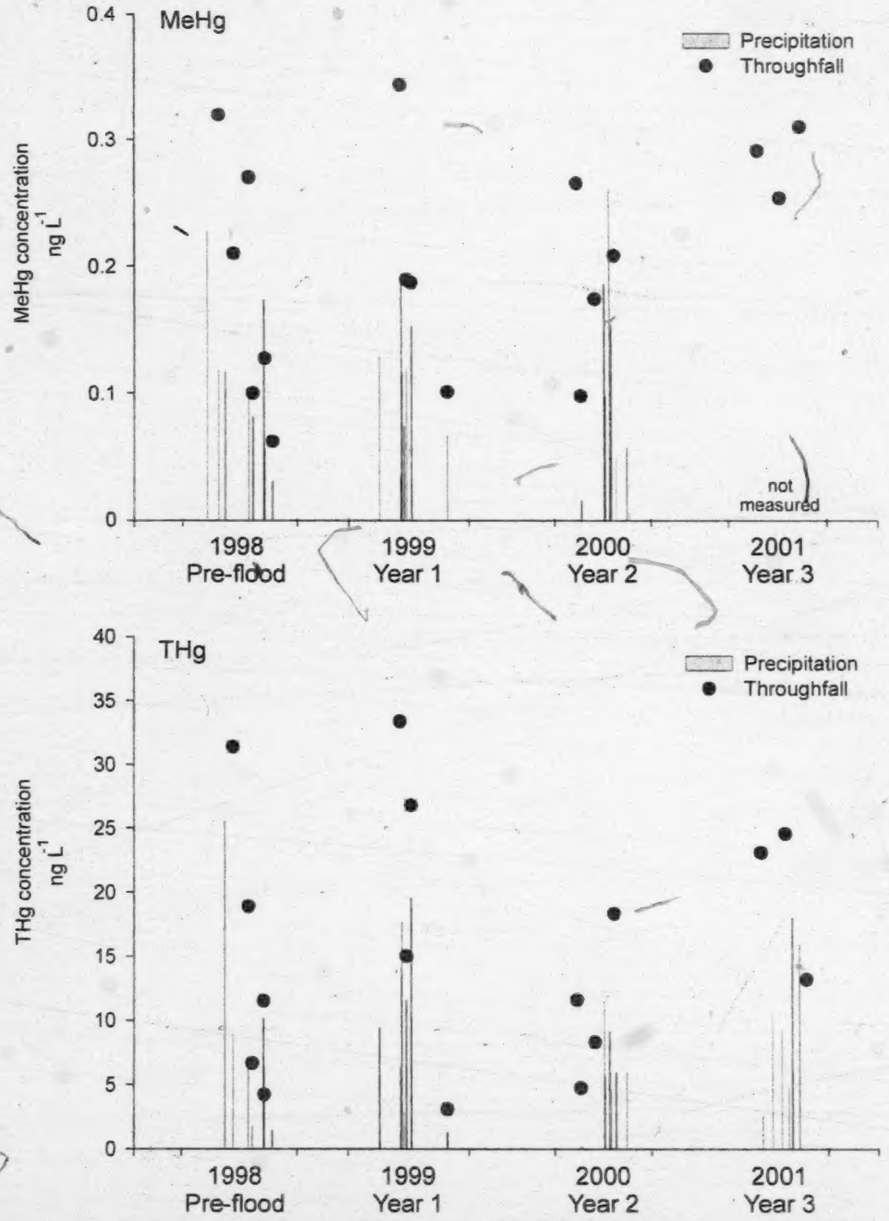
Appendix 11. Locations of artificial substrates for sampling periphyton from the Medium C reservoir.


RB
Boulder
RA
RC
W
D
M
Limit of Flooding
0
10
20
30
40
50 Metres
Experimental Lakes Area
FLUDEX
Site 3
LEGEND
Weir
Drain
Inflow
Periphyton Artificial Substrates
Raft (A-C)
Flooding Extent
Dam Wall
Transport Paths
Contour (metres)
Dike
Reference: Samot 2001
By: C.E. (June 2002)

Appendix 12. Locations of artificial substrates for sampling periphyton from the Low C reservoir.



Appendix 13. Mass (kg ha^{-1}) and carbon (kg C ha^{-1}) in litterfall falling into each FLUDEX reservoir in the first three years post flood.



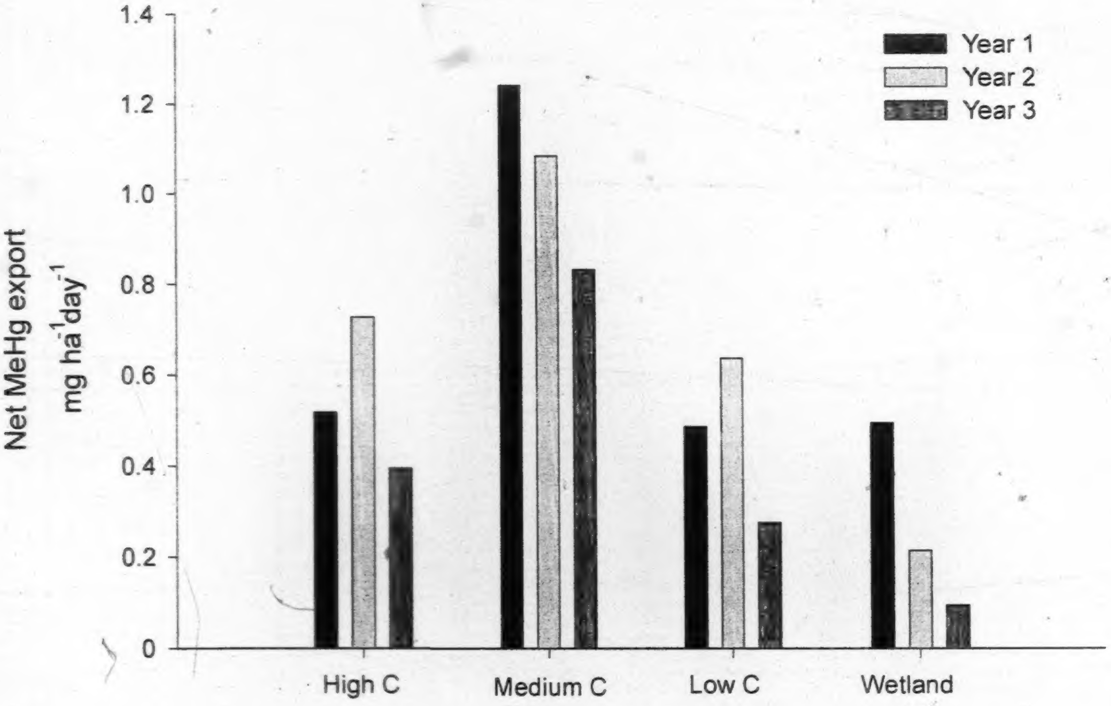
Appendix 14. Methylmercury (MeHg) and total mercury (THg) concentrations (ng L^{-1}) in precipitation and throughfall at the Experimental Lakes Area (1998-2001).

Appendix 15. Average methylmercury (MeHg) and total mercury (THg) concentrations (ng L^{-1}±one standard error) for wet deposition in the open and throughfall (volume weighted) and in direct runoff.

	Year 1 (1999)		Year 2 (2000)		Year 3 (2001)	
	ng L^{-1}	n	ng L^{-1}	n	ng L^{-1}	n
Wet deposition in the open						
MeHg	0.128 ± 0.020	7	0.071 ± 0.018	6	Not measured	5
THg	13.21 ± 2.71	7	3.87 ± 0.72	6	9.24 ± 2.47	6
Throughfall						
MeHg	0.215 ± 0.031	7	0.153 ± 0.044	4	0.295 ± 0.017	3
THg	24.70 ± 4.81	6	7.71 ± 2.46	4	19.17 ± 3.57	3
Direct runoff						
MeHg	0.039 ± 0.013	6	0.025 ± 0.010	2	0.104 ± 0.013	3
THg	9.11 ± 0.92	8	10.21 ± 1.21	2	8.36 ± 0.73	3

Appendix 16. Average methylmercury (MeHg) and total mercury (THg) concentrations (ng L^{-1}±one standard error) in bottom waters of upland reservoirs in September of each year.

	Year 1		Year 2		Year 3	
	ng L^{-1}	n	ng L^{-1}	n	ng L^{-1}	n
High C						
MeHg	1.78 ± 0.83	3	2.66 ± 2.03	3	0.54 ± 0.04	3
THg	13.29 ± 9.46	3	2.61	1	2.48 ± 0.33	3
Medium C						
MeHg	0.73 ± 0.21	3	0.44 ± 0.04	3	0.37 ± 0.08	3
THg	3.19 ± 0.37	3	1.87 ± 0.10	3	1.70 ± 0.31	3
Low C						
MeHg	0.53 ± 0.01	3	0.19 ± 0.03	3	0.19 ± 0.03	3
THg	3.26 ± 1.35	3	2.23 ± 0.41	3	1.13 ± 0.35	3



Appendix 17. Methylmercury exports (mg MeHg ha^{-1}) from the upland and wetland reservoirs when surface water temperatures were 15°C or higher (June 15 – August 31).

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Education

Ph.D., Environmental Biology and Ecology, Department of Biological Sciences, University of Alberta, 2003

"Impacts of reservoir creation on the biogeochemical cycling of methylmercury in boreal forest uplands." Supervisor: Dr. V. St. Louis

M. Sc., University of Manitoba, Department of Entomology, 1996.

"Bioaccumulation of methylmercury by aquatic insects and fish at the Experimental Lakes Area." Supervisor: Dr. David Rosenberg

B. Sc., University of Manitoba, Departments of Chemistry and Microbiology, 1992; Major in Biochemistry

Scholarships and awards

Name of award	Institution	Value	Date held
Research awards			
Province of Alberta Graduate Fellowship	University of Alberta	$10700CDN	2002-2003
Graduate Research Assistantship	University of Alberta	$15320CDN	1999; 2001
C/BAR Award[1]	C/BAR[1] Institute	$2500CDN	1999-2001
D. Alan Birdsall Memorial Scholarship	University of Alberta	$6500CDN	1999-2000
Runner up- Best Research Day Seminar	University of Alberta		1999
Teaching assistantship (competitive award based on academic standing)			
Graduate Teaching Assistantship	University of Alberta	$11555CDN	1999-2002
Travel awards			
Travel Award	ASLO[2]	$200US	2002
J. Gordin Kaplan Travel Award	University of Alberta	$500CDN	2001
Department of Biological Sciences Travel Award	University of Alberta	$300CDN	2001

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[1] Circumpolar/Boreal Alberta Research

[2] American Society of Limnologists and Oceanographers

Publications

Refereed Publications:

1. **Hall, B.D.**, V.L. St. Louis, K.R. Rolfhus, R.A. Bodaly, K.G. Beaty, M.J. Paterson, and K.A. Peech Cherewyk. Impacts of reservoir creation on the biogeochemical cycling of methylmercury in boreal forest uplands. Submitted to *Ecosystems*; June 2003
2. C.J.D. Matthews, E.M. Joyce, V.L. St. Louis, S.L. Schiff, J. J. Venkiteswaran, **B.D. Hall**, R.A. Bodaly, and K. G. Beaty. Carbon dioxide (CO_2) and methane (CH_4) production in small reservoirs flooding upland boreal forests. Submitted to *Ecosystems*; January 2003.
3. **Hall, B.D.**, V.L. St. Louis, and R.A. Bodaly. in press. The stimulation of methylmercury production by decomposition of flooded birch leaves and jack pine needles. *Biogeochemistry*.
4. St Louis, V.L., J.W.M. Rudd, C.A. Kelly, **B.D. Hall**, K.R. Rolfhus, and K.J. Scott. 2001. The forest canopy as a possible important input of methyl mercury and inorganic mercury to boreal ecosystems. *Environ. Sci. Technol.* 35:3089-3098. *Note: This publication was chosen by the editors to be featured in Environ. Sci. Technol. Online News.*
5. **Hall, B.D.**, D.M. Rosenberg, and A.P. Wiens. 1998. Methyl mercury in aquatic insects from an experimental reservoir. *Can. J. Fish. Aquat. Sci.* 55:2036-2047.
6. **Hall, B.D.**, R.A. Bodaly, R.J.P. Fudge, and J.W.M. Rudd. 1997. Food as the dominant pathway of methylmercury uptake by fish. *Water Air Soil Pollut.* 100:13-24.
7. Bodaly, R.A., V.L. St. Louis, M.J. Paterson, R.J.P Fudge, **B.D. Hall**, D.M. Rosenberg, and J.W.M. Rudd. 1997. Bioaccumulation of mercury in the aquatic food chain in newly flooded areas. *In:* Mercury and its effects on environment and biology. H. Sigel and A. Sigel (eds.) Marcel Dekker, New York, N.Y. pg. 259-287.
8. Malley, D.F., A.R. Stewart, and **B.D. Hall**. 1996. Uptake of methylmercury by the floater mussel, *Pyganodon grandis* (Bivalvia, Unionidae) caged in a flooded wetland. *Environ. Toxicol. Chem.* 15:928-936.

In Preparation:

9. **Hall, B.D.**, and V.L. St. Louis. Methylmercury and total mercury in plant litter decomposing in upland forests and flooded landscapes.

Invited conference communications

1. **Hall, B.D.**, V.L. St. Louis, R.A. Bodaly, J.W.M. Rudd, and C.A. Kelly. Increases in exports of methylmercury from reservoirs experimentally created over upland forest and wetland landscapes. American Society of Limnology and Oceanography, Salt Lake City, Utah. 9-14 February 2003.
2. **Hall, B.D.**, R.A. Bodaly, M.J. Paterson, D.M. Rosenberg, J.W.M. Rudd, C.A. Kelly, and V.L. St. Louis. The Experimental Lakes Area Reservoir Project: An example of whole ecosystem manipulation to study the effects of reservoir creation. Ecological Monitoring Assessment Network 2nd Annual Meeting. Halifax, Nova Scotia. 20 January 1996.

Conference communications

1. **Hall, B.D.**, R.A. Bodaly, K.G. Beaty, D.L. Findlay, L.L. Hendzel. J.P. Hurley, A.R. Majewski, C.J.D. Matthews, M.J. Paterson, K.A. Peech, K.R. Rolfhus, V.L. St. Louis, S.L. Schiff, J. Venkiteswaran. Mercury cycling and greenhouse gas fluxes from flooded boreal forest uplands: The FLUDEX (Flooded Uplands Dynamics Experiment) project at the Experimental Lakes Area, northwestern Ontario. American Society of Limnology and Oceanography, Victoria, British Columbia. June 10-14, 2002.
2. **Hall, B.D.**, V.L. St. Louis, R.A. Bodaly, and K.G. Beaty. Impacts of reservoir creation on the biogeochemical cycling of methylmercury in boreal forest uplands. 6th International Conference on Mercury as a Global Pollutant. Minamata, Japan. 15-19 October 2001.
3. **Hall, B.D.**, V.L. St. Louis, and R. A. Bodaly. Methylmercury production in flooded jack pine needles and birch leaves. 6th International Conference on Mercury as a Global Pollutant. Minamata, Japan. 15-19 October 2001.
4. **Hall, B.D.**, V.L. St. Louis, R.A. Bodaly, and K.G. Beaty. Methylmercury production in flooded forest uplands. Society of Environmental Toxicology and Chemistry Annual Meeting (SETAC) 21st Annual Meeting. Nashville, Tennessee. 12-16 November 2000.
5. **Hall, B.D.**, V.L. St. Louis, and R.A. Bodaly. Impact of reservoir creation on the biogeochemical cycling of methylmercury in boreal forest uplands. Mercury in the Environment Speciality Conference. Minneapolis, Minnesota. 15-17 September 1999.

6. **Hall, B.D.**, V.L. St. Louis, and R.A. Bodaly. Impacts of reservoir creation on the biogeochemical cycling of MeHg in boreal forest uplands: Pre-flood results. Society of Canadian Limnologists Annual Meeting. Edmonton, Alberta. 9-11 January 1999.
7. **Hall, B.D.**, D.M. Rosenberg, A.P. Wiens, R.A. Bodaly, C.A. Kelly, and J.W.M. Rudd. Bioaccumulation of methylmercury by aquatic insects in an experimental reservoir. 4th International Conference on Mercury as a Global Pollutant. Hamburg, Germany. 4-8 August 1996.
8. **Hall, B.D.**, R.A. Bodaly, R.J.P. Fudge, and J.W.M. Rudd. Food as the dominant pathway of methylmercury uptake by fish. 3rd International Conference on Mercury as a Global Pollutant. Whistler, British Columbia. 10-14 July 1994.

Contributed conference communications

1. Paterson, M.J., K.A. Peech, R.A. Bodaly, **B.D. Hall**, A. Majewski, R.J.P. Fudge, M. Pinsonneault, D.M. Rosenberg, J.W.M. Rudd, L. Wesson, A.P. Wiens. Methylmercury in new reservoirs: The Flooded Uplands Dynamics Experiment. American Society of Limnology and Oceanography, Salt Lake City, Utah. 9-14 February 2003.
2. Peech, K.A., M.J. Paterson, Bodaly, R.A. and **B.D. Hall**. Methylmercury in the food webs of new reservoirs: the Flooded Uplands Dynamics Experiment. American Society of Limnology and Oceanography, Victoria, British Columbia. June 10-14 2002.
3. Rolfus, K.R., J.P. Hurley, **B.D. Hall**, and D.P. Krabbenhoft. Mercury fluxes from inundated soils of upland boreal reservoirs. International Association of Great Lakes Research. Winnipeg, Manitoba. 6-9 June 2002.
4. Rolfus, K.R., J.P. Hurley, **B.D. Hall**, and D.P. Krabbenhoft. The response of soil/water mercury fluxes to periodic inundation of upland boreal forest reservoirs. 6th International Conference on Mercury as a Global Pollutant. Minamata, Japan. 15-19 October 2001.
5. Paterson, M.J., R.A. Bodaly, K.A. Peech, R.J.P. Fudge, **B.D. Hall**, A. Majewski, D. Rosenberg, A.P. Wiens. Methylmercury in the food webs of experimental reservoirs and natural lakes at the Experimental Lakes Area, Canada. 6th International Conference on Mercury as a Global Pollutant. Minamata, Japan. 15-19 October 2001.
6. R.A. Bodaly, R.J.P Fudge, **B.D. Hall**, M.J. Paterson, K.A. Peech, K.R. Rolfus, and V.L. St. Louis. Methyl mercury production and bioaccumulation in three experimental reservoirs that flooded boreal forest uplands. Workshop on the fate, transport, and transformation of

mercury cycling in aquatic and terrestrial environments. West Palm Beach, Florida. 8-10 May 2001.

7. St. Louis V.L., J.W.M. Rudd, C.A. Kelly, **B.D. Hall**, K.R. Rolfus, R.A. Bodaly, K.G. Beaty, and S.E. Lindberg. An overview of mercury cycling in the boreal ecosystem. Workshop on the fate, transport, and transformation of mercury cycling in aquatic and terrestrial environments. West Palm Beach, Florida. 8-10 May 2001.
8. St. Louis, V.L., J.W.M. Rudd, C.A. Kelly, **B.D. Hall**, K.R. Rolfus, K.J. Scott, and S.E. Lindberg. The forest canopy as a possible important input of methyl mercury and inorganic mercury to boreal ecosystems. Society of Environmental Toxicology and Chemistry Annual Meeting (SETAC) 21st Annual Meeting. Nashville, Tennessee. 12-16 November 2000.
9. Rolfus, K.R., R.A. Bodaly, R.J.P. Fudge, **B.D. Hall**, V.L. St. Louis, D. Huebert, D.P. Krabbenhoft, J.P. Hurley, K. Peech, and M.J. Paterson. The upland flooding experiment: Assessing the impact of reservoir creation on the biogeochemical cycling of mercury in boreal forest uplands. CPANS/PNWIS/A&WMA Air toxics 2000 Conference, Banff, Alberta. 9-12 April 2000.
10. St. Louis, V.L., J.W.M. Rudd, C.A. Kelly, **B.D. Hall**, K.R. Rolfus, and K.J. Scott. The forest canopy as a possible important input of methyl mercury and inorganic mercury to boreal ecosystems. CPANS/PNWIS/A&WMA Air toxics 2000 Conference, Banff, Alberta. 9-12 April 2000.
11. M.J. Paterson, Bodaly, R.A., R.J.P. Fudge, **B.D. Hall**, D.M. Rosenberg, J.W.M. Rudd, V.L. St. Louis, L. Wesson, and A.P. Wiens. Mercury dynamics in the food web of an experimental peatland reservoir. Mercury in the Environment Speciality Conference. Minneapolis, Minnesota. 15-17 September 1999.
12. Bodaly, R.A., M.J. Paterson, D.M. Rosenberg, R.J.P. Fudge, **B.D. Hall**, A.P. Wiens, J.W.M. Rudd, and V.L. St. Louis. Increases in mercury in fish and invertebrates in an experimental boreal reservoir. 4th International Conference on Mercury as a Global Pollutant. Hamburg, Germany. 4-8 August 1996.
13. Rudd, J.W.M., C.A. Kelly, R.A. Bodaly, V.L. St. Louis, D.M. Rosenberg, **B.D. Hall**, and R. Harris. Production, transport and bioaccumulation of methylmercury in natural and experimentally flooded boreal ecosystems. Mercury Pollution in the Upper Great Lakes Region. Minneapolis, Minnesota. 9 June 1995.

Invited seminars

1. **Hall, B.D.** V.L. St. Louis, R.A. Bodaly, and K.G. Beaty. Methylmercury production in flooded forest uplands. Experimental Lakes Area. 8 August 2001.
2. St. Louis, V.L., **B.D. Hall**, and C.J.D. Matthews. Fluxes of mercury and greenhouse gases from reservoirs as determined by whole ecosystem experimentation. Department of Biological Sciences, University of Alberta. 14 March 2000.
3. **Hall, B.D.**, D.M. Rosenberg, A.P. Wiens, R.A. Bodaly, C.A. Kelly, and J.W.M. Rudd. Bioaccumulation of methylmercury by aquatic insects in an experimental reservoir. Experimental Lakes Area. August 1994.
4. **Hall, B.D.**, R.A. Bodaly, R.J.P. Fudge, and J.W.M. Rudd. Food as the dominant pathway of methylmercury uptake by fish. Experimental Lakes Area. August 1993.

Teaching experience

Laboratory Teaching Assistantship

- Introductory Biology; Winter term 1999, 2000, 2001, 2003. Instructed first year undergraduate students on a wide range of biology concepts
- Freshwater Ecology; Fall term 2002. Advised and evaluated senior undergraduate students on oral presentations given on publications chosen from ecology literature

Student Evaluation / Marker

- Introductory Ecology; Winter term 2001
- Pollution Biology; Winter term 1998, Fall term 2002
- Introductory Entomology; Winter term 1996

Guest Lecturer

- Biogeochemistry; Winter term 2002
- Pollution Biology; Winter term 1998

Undergraduate Students Supervised

- Megan Puchniak, 2001
- Michelle Pinsonneault, 2000
- Pauline Gerrard, 1994
- Sheena Majewski, 1993
- John Embury, 1992

Training

- Completion of the University of Alberta Teaching Program. The primary objectives of the UT Program are to allow graduate students to develop ethical, philosophical, and practical skills for careers in post-secondary teaching. The UT Program requirements include pedagogical, practicum, and documentation components.

  Teaching Dossier available upon request

Employment experience

Registered Professional Biologist - Consultant

Rescan Environmental Services, Vancouver, British Columbia (June, 1996-December 1997)

- designed and implemented several environmental monitoring programs for mining companies with properties in British Columbia and the Northwest Territories. This work also involved remote fieldwork, data collection and analysis, management of sub-consultants, as well as writing, editing, and producing several scientific reports.

Laboratory Analyst

University of North Carolina at Charlotte, Winnipeg, Manitoba (May 1996)

- methyl and total mercury analysis of soil profile samples using organic extraction, aqua regia digestion, and atomic absorption spectrophotometry.

Field Research Technician

Fraser River Action Plan (FRAP), Department of Environment, Government of Canada, Vancouver, British Columbia (October 1995)

- established base line knowledge for the Fraser River biomonitoring program. An intensive helicopter survey of the main stem and tributaries entailed the collection of water chemistry samples from remote mountain streams, collection of aquatic insects using kick nets and leaf pack methods, and quantitative sampling of epiphytic periphyton.

Professional Associations

- American Society of Limnology and Oceanography (2002-present)
- Alberta Society of Professional Biologists (Student Member 1999-2000)
- Society of Canadian Limnologists (1998-1999)
- Association of Professional Biologists of British Columbia (1997-1998)

Other scientific / creative activities

Workshops attended

Annual Flooded Upland Dynamics Experiment (FLUDEX) Workshop

- 14-16 April 2002, Madison, Wisconsin
- 7-10 April 2001, Jasper, Alberta
- 7-9 April 2000, Jasper, Alberta
- 29-31 March 1999, Winnipeg, Manitoba

Hydro Utilities Mercury and Greenhouses Gases in Reservoirs

- 9-12 February 2002, Montreal, Québec
- 17-18 October 1999, St. John's, Newfoundland
- 7-9 November 1998, New Westminster, British Columbia
- 5-6 October 1997, Winnipeg, Manitoba
- 17-18 March 1996, Montreal, Québec
- 14-15 March 1994, Winnipeg, Manitoba

Methylmercury in Freshwater Foodwebs: Current Knowledge, Methods and Models. 2-4 May 1995, Trout Lake Limnological Station, Wisconsin

Discussion groups

- Participation in a weekly senior undergraduate/graduate student discussion course to aid in discussion and clarification of topics in biogeochemistry (January - March, 2001)
- Participation in a weekly graduate student and faculty member group that critiqued new papers in the field of ecology (1998-2000)

Certificates

- PADI SCUBA Divemaster;
- PADI Emergency First Aid and CPR;
- Radiation Safety Course;
- Class 7 (Radioactive) Receiver;
- Canadian Coast Guard Pleasure Craft Operator.